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TRITIUM CONTAMINATION IN PARTICLE ACCELERATOR OPERATION

Donald O. Nellis, et al

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TRITIUM CONTAMINATION IN PARTICLE ACCELERATOR OPERATION

by

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ABSTRACT

Tritium-metallic targets are frequently used in many particle accelerators for the production of high-energy neutrons. The purpose of this study was to investigate the problems and hazards associated with the use of these tritium targets so that appropriate steps can be taken to minimize the potential radiological health hazards. Particular emphasis was placed on the operation of Cockcroft-Walton neutron generators, since these constitute a high percentage of such particle accelerators which utilize the tritium-metallic targets.

This study showed that bombardment of tritium targets results in the loss of about 1 tritium atom for each bombarding deuteron, and that most of the tritium is in a gaseous form when it leaves the target. It was also determined that a relatively small amount of the tritium released from the target actually remains within the accelerator itself. Most of the tritium lost by a bombarded target is trapped in the elements of an ion pump or released through the vacuum system exhaust.

Representative products and manufacturers are named for identification only, and listing does not imply endorsement by the Public Health Service and the U.S. Department of Health, Education, and Welfare.

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TRITIUM CONTAMINATION IN PARTICLE ACCELERATOR OPERATION

INTRODUCTION

Tritium-metallic targets are now commonly used for the production of neutrons in hundreds of particle accelerators (neutron generators) in the United States and throughout the world. Gaseous targets of tritium are also used, but such use is restricted to a relatively small number of laboratories. In a few installations, tritium is fed into an ion source and the emergent tritons are accelerated and used in nuclear reaction studies. In all of these cases, tritium represents a potential hazard to accelerator operators and users. Even in cases of sealed-tube neutron generators, emergencies may arise through accidential breakage of a tube.

A typical tritium-metallic target contains about 5 curies of activity and will usually be depleted at a rate of tens to hundreds of millicuries/hour of bombardment (or even at a rate of curies/hour in a very high-current machine). A gaseous tritium cell will probably contain at least a few cubic centimeters of tritium at a concentration of 2.6 curies/cc. An ion source for triton production may well pass approximately a curie/hour of tritium. A sealed tube generally contains 5 to 10 curies of tritium, either absorbed within the tube or in the gaseous discharge. Therefore, the amount of tritium involved in any of the equipment mentioned above lies in the curie region and is potentially dangerous.

In order to minimize the hazards associated with tritium, it is necessary to know where it may accumulate in a system, how rapidly it is expelled by pumps, and what procedures to follow in case of accidental release of a large amount of tritium into a laboratory. This report describes experimental investigations of certain tritium hazards and presents conclusions regarding safe operating procedures.

Tritium accountability studies with a Cockcroft-Walton type accelerator (neutron generator) have received major attention. This is

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because (1) there are many more neutron generators in use for routine production of high-neutron fluxes; (2) the hazards of sealed tube operation usually occur only if the tube is accidentally broken by the user; and (3) the fundamental studies on the mechanisms by which tritium is lost from a target are most readily evaluated with the high bombarding currents produced by low-energy generators (approximately 150 keV). However, studies have also been performed with other equipment, and it is believed this report will answer many questions relating to tritium hazards around particle accelerators in general.

SECTION I. DETERIORATION OF TRITIUM-METALLIC TARGETS

It is well known that tritium-metallic targets show deterioration with respect to neutron output when they are placed under ion bombardment, but the reasons for this are not known. One may assume that tritium is actually driven from the target by the bombarding ions. On the other hand, Akers and Banta (*1*) state "Many experimenters insist that loss of activity by the target is due to cracking of the organic vapors in the vacuum system, leading to the deposition of a layer of elemental carbon on the face of the target." They conclude that this is not true, pointing out that targets tend to deteriorate similarly in machines equipped with oil diffusion pumps and with solid state pumps. However, it is extremely difficult to rid any vacuum system of organic vapors (even with solid state pumping), and it has been attempted, in some of the work reported here, to obtain direct evidence concerning the amount of tritium that actually leaves a target under bombardment. This amount defines the magnitude of the tritium accountability problem—if tritium were simply buried or driven into the target by the bombarding ions, then the tritium hazards would obviously be confined to it alone.

It is well established that some tritium does leave the target. Akers and Banta (*1*) found indications that both the diffusion pump and the forepump used in their work contain "multicurie amounts of tritium." On the other hand, Broerse (*2*), in studies with 400-keV deuteron bombardments of tritium titanium targets, states that the tritium activity of the diffusion pump oil was only 0.3 μCi/cc in a 100 cc total volume. These differences in estimated contamination of pump oil may be ascribed to differing conditions and histories of bombardment and to time of withdrawal of samples. In any event, it was clear that more investigation should be made of this point.

The mechanism of the loss of tritium by a target is also of fundamental interest. Knowledge on this point could lead to development of targets that show improved performance with respect to neutron output and present a reduced tritium hazard. Although it is believed

by Akers and Banta (*1*) that "the decay of neutron yield with time is due to displacement of the tritium by bombarding deuterons," it was found by Broerse (*2*) that a deuterium-metallic target showed decreased neutron output under deuteron bombardment. Thus it appears that the tritium that leaves a target is not fully replaced by bombarding deuterons.

An account is presented herein of the target deterioration studies. Tritium loss was measured directly, and some information was gained on the mechanism of loss. A general definition for a "figure of merit" was derived which should be useful in describing performance of tritium targets.

A. Mapping of T/Ti Targets

The amount of tritium lost by a target is most conveniently measured by determining its initial activity and then subtracting from this its activity after a monitored bombardment. Three methods were used for measuring the tritium activity and correlations have been made between them. One of these methods (based on the T(p,n) reaction) yields absolute results, and these results may be compared with the tritium uptake measurements supplied by the target manufacturer.

The measurement of tritium concentration at representative areas over the face of a target is referred to as "mapping the target." An activity map consists of a series of contour lines, each of which represents a specific tritium activity. Contour maps made before and after target bombardment may then be compared in order to determine the amount of tritium lost through the bombardment. Each method of target mapping and the correlations between them are described below.

MEASUREMENT OF T/Ti K X-RAY YIELD

The beta rays from the tritium in a T/Ti target will excite characteristic Ti K X rays. The intensity of these X rays is proportional to the concentration of tritium in the target (assuming that X-ray absorption may be disregarded in these thin targets). A target may be mapped by observing the Ti K X rays from each of several selected areas of the target. Such observations were possible through the use of a target handling device which allows both rotational and translational motion of targets with respect to a fixed collimating aperture and en-

ables the experimenter to locate any chosen area within 0.5 mm in either direction. Figure 1 depicts the positioning device in an open position, and figure 2 shows it mounted above the proportional counter which was used for X-ray detection. The output of the counter was fed to a multichannel analyzer and a window was set on those counts of interest. Figure 3 shows the 4.5-keV Ti X-ray peak which was observed in a typical run. In mapping each target, 25 selected locations were separately positioned and the X-ray intensity observed.

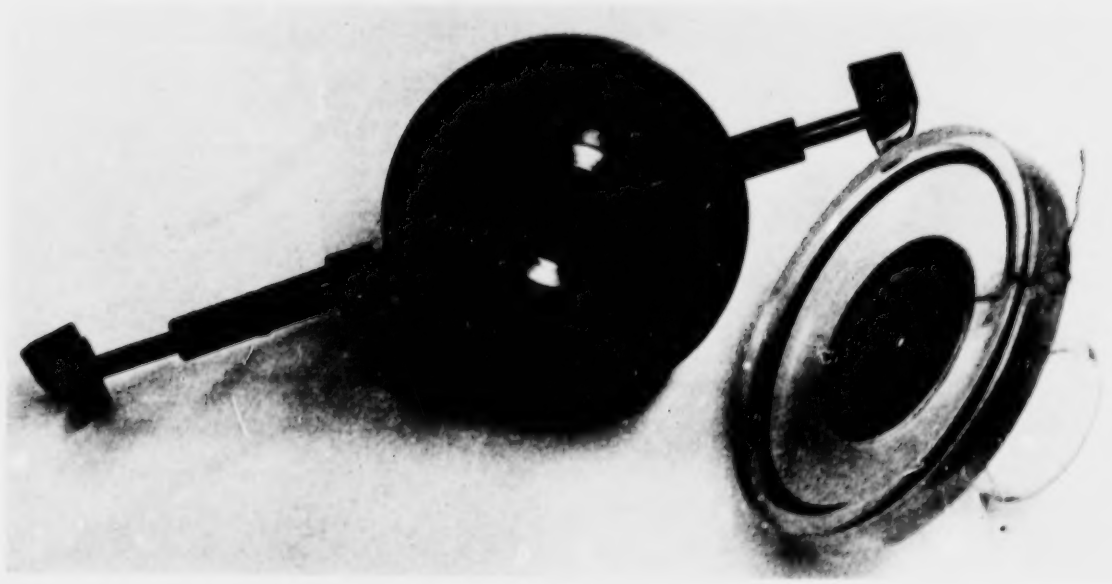

Figure 1. Target positioning device in open position. The T/Ti target is mounted in the center of the assembly. It may be rotated or moved laterally by two knobs allowing any portion of the target to be centered in the device. The cover plate at the right supports a collimated holder through which the Ti K X rays emitted by the target may pass. This same positioning device permitted systematic proton bombardment of selected areas of the target.

MAPPING BY THE T(p,n) REACTION

Tritium concentration may be determined also by its reaction with energetic protons. The reaction is $T+p \rightarrow {}^3He+n$, where the symbols T, p, and n stand respectively for triton, proton, and neutron. The reaction has a threshold value of 1.02 MeV and is readily observed at energies above that.

The target positioning device described in figures 1 and 2 was connected through an adapter to the drift tube of a Van de Graaff accelerator and used to hold a tritium target in position for proton bombard-

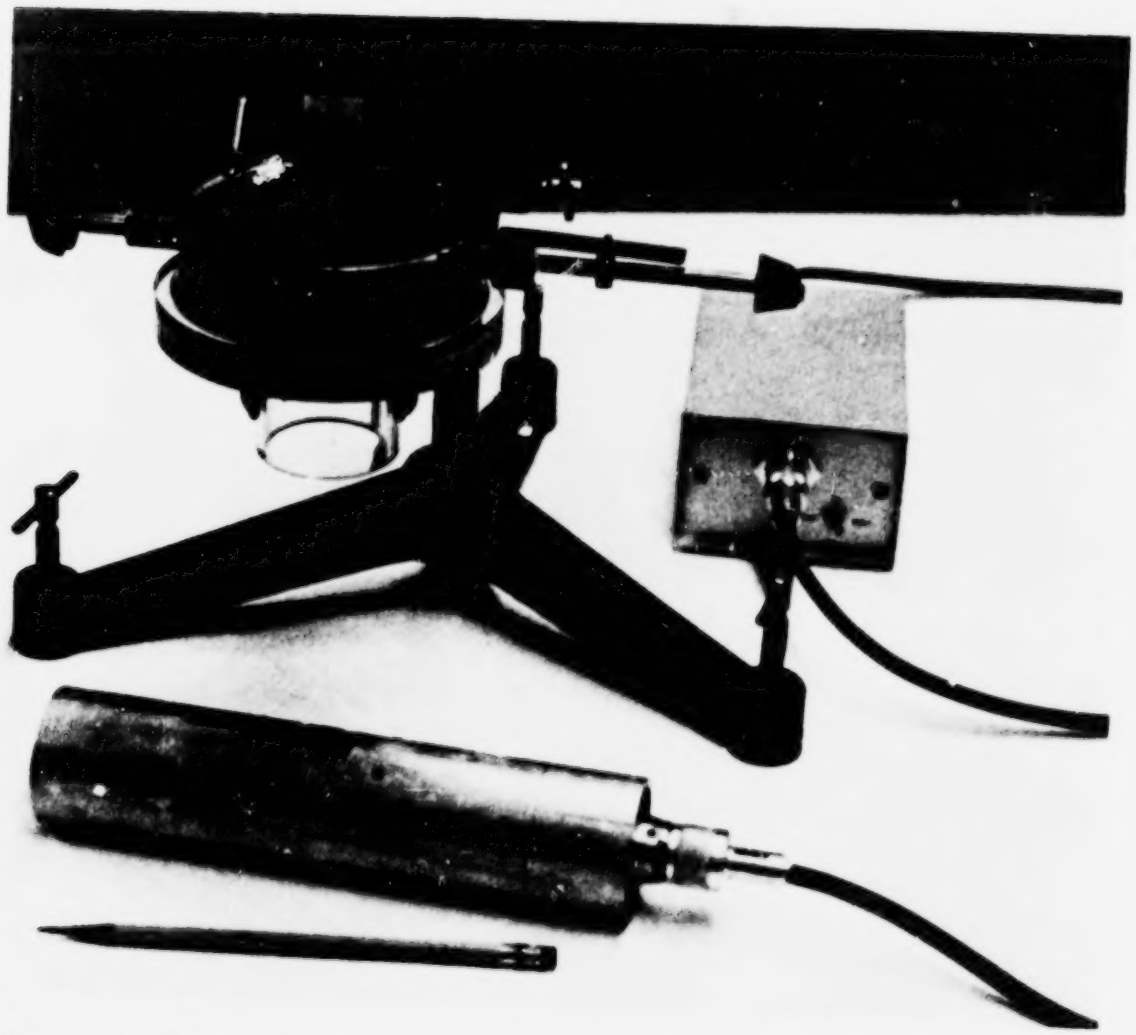

Figure 2. Target positioning device mounted above a proportional counter. The counter was used to detect the Ti K X rays from various positions of the target. Maps of these data yield contour lines of specific tritium concentration in the target.

ment of selected areas. In addition to the ⅛-inch tantalum collimator in the positioning device, another ⅛-inch collimator was placed 3 inches away in order to further define the beam. The mean proton energy was 2.67 MeV, yielding neutrons of 1.88-MeV energy at 0°. The targets were mapped along six diameters, each 30° apart, for a total of 25 measurements on each target. The neutrons produced from each of the target positions were measured with a calibrated polyethylene long counter placed 1 meter from the target at 0° to the incident beam. Calibration of the long counter had been determined by comparison with a proton recoil telescope. Following the end of the bombardments, one of the targets was reversed and the copper backing bombarded with protons to check on possible interference effects from the $^{65}Cu(p,n)$ ^{65}Zn reaction, which has a threshold near 2.1 MeV. The yield from this

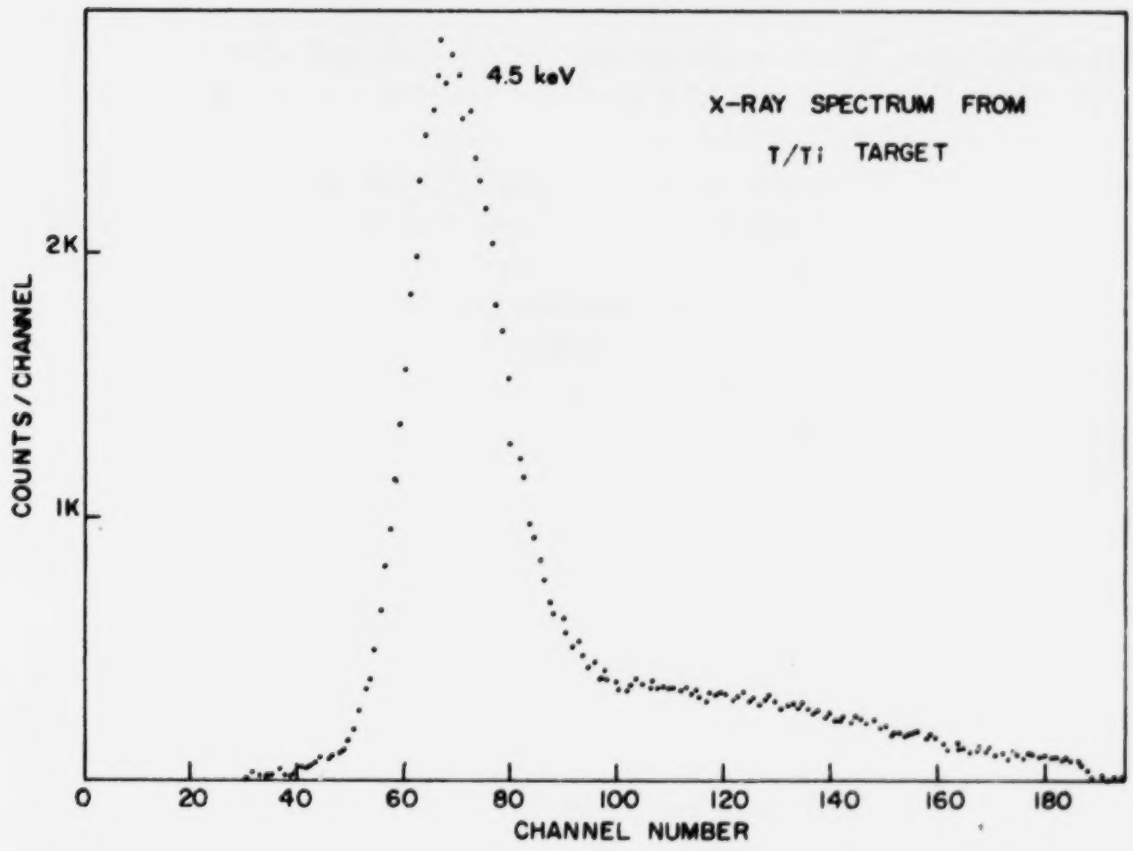

Figure 3. Map of the X-ray spectrum obtained from a T/Ti target. The proportional counter shown in figure 3 was used as a detector. The characteristic K X ray from titanium is the only one which is apparent. (The characteristic K X ray from copper has an energy of 8.05 keV; there is no indication of it.)

reaction proved to be less than 1 percent of the counts observed from the tritium side and was therefore neglected.

From the known cross section for the T(p,n) reaction at a mean bombarding energy of 2.67 MeV and from the measurement of neutron yield produced by a monitored bombardment, the absolute amount of tritium in the bombarded region may be readily calculated.

AUTORADIOGRAPHS OF TRITIUM TARGETS

Autoradiographs of T/Ti targets are made by simply placing the target gently against photographic film for a brief exposure. Several common films (including regular roll film, dental X-ray film, and Polaroid, normal and reversal, with exposure times from ½ to 30 seconds) have been used, but the autoradiographs used in subsequent densitometer measurements were made with single-coated industrial X-ray film, Type R, with exposures generally ranging from ½ to 3 minutes.

CORRELATIONS BETWEEN RESULTS OF MAPPING BY VARIOUS PROCEDURES

Absolute measurement of tritium concentration may be accomplished by the T(p,n) measurements, as mentioned above, but relatively few laboratories are in a position to carry out this procedure. It is also an expensive method. However, T(p,n) studies have been used to check the tritium uptake measurements which were supplied by the manufacturer of the targets. (The manufacturer, on the basis of tritium uptake measurements, rates the strength of each target in curies of activity.) The results of these checks are given below. It is also important to determine whether the results of mapping by measurement of the Ti K X rays and by autoradiographs may be directly correlated with the T(p,n) results. Typical correlations are presented on page 9.

Comparison of Absolute Measurements of Tritium Concentration of Targets

The tritium concentration of several targets determined from the T(p,n) measurements at Texas Nuclear Corp. have been compared with the quoted target concentration as specified by the manufacturer (table 1). The target concentration as specified by the manufacturer is calculated from the pressure decrease in a calibrated volume as the tritium is absorbed by the titanium. Considering the accuracy of the instruments used and the time difference between the two measurements during which some of the targets might have evolved some of the absorbed tritium, the agreement is satisfactory.

Table 1. Comparison of tritium activity based on uptake of tritium gas versus results of T(p,n) measurements

Target	Manufacturer (atoms $\times 10^{19}$)	Measured by T(p,n) (atoms $\times 10^{19}$)	Ratio of measured values to manufacturer's values
L–95	7. 5	5. 8	0. 77
L–96	7. 3	7. 6	1. 05
L–97	7. 6	6. 3	. 83
L–98	10. 2	9. 1	. 90
L–99	6. 5	4. 8	. 74

Through this T(p,n) work—and also in all other mapping studies—it was possible to determine the variation in uniformity of tritium concentration on the surface of targets. The variations are usually gradual and amount to approximately ± 10 percent in a typical case.

Correlation Between T(p,n) Mapping and X-ray Mapping

The ratios between the Ti K X-ray yield and the neutron yield from the T(p,n) reaction for a specific area of any target proved to be quite consistent, as shown in table 2. In addition, the average ratio was essentially the same. The ratios obtained apply for the particular geometry used in the sets of X-ray measurements. A suitable "normalization" factor could be extracted for use in any geometry, and thereafter absolute activity of thin T/Ti targets could be made by observation of the X rays.[1] The ratio (with an estimated error of 10 percent) is given in the second column of table 2, and the number of readings (of a total of 25 on each target) falling outside the 10-percent error is given in the third column. It should be noted that the number of these readings with errors exceeding 10 percent decreased in the postbombardment mappings in relation to the prebombardment mappings. This resulted from a minor improvement in the experimental setup for the measurement of the Ti X ray. Further refinements in the method of target positioning for Ti X-ray measurements would most likely lead to even more consistent ratios. In view of these results, it was concluded that X-ray mapping offers a practical means for determining the tritium distribution in targets of this type.

Table 2. Ratio of neutron yield by T(p,n) reaction to yield by Ti K X-ray mapping

Target	Prebombardment		Postbombardment	
	Ratio [a]	Number exceeding 10 percent [b]	Ratio [a]	Number exceeding 10 percent [b]
L–95	2.72 ± 10 percent	1	2.65 ± 10 percent	0
L–95	2.63 ± 10 percent	1	2.60 ± 10 percent	1
L–97	2.68 ± 10 percent	1	2.58 ± 10 percent	1
L–98	2.74 ± 10 percent	2	2.70 ± 10 percent	0
L–99	2.60 ± 10 percent	1	2.62 ± 10 percent	0

[a] Ratios of neutron to X-ray yield.
[b] This refers to the number of mapped positions of the 25 mapped on each target where the ratio exceeds the 10-percent error quoted.

[1] The appropriate normalization factor may be determined by observing the X-ray yield from the target as a whole, with the detector placed a good distance away from the target. From the known strength of the target, the ratio of the number of X-ray counts/sec/Ci of target activity is immediately established for the particular geometry employed. Calculations pertinent to other geometries may then be readily made.

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Correlation Between X-ray Mapping and Autoradiographs

In order to compare the mapping results obtained through Ti K X-ray measurements and by means of an autoradiograph, a target (designated as L–44) was examined by both methods. It was desired to examine a target that had definite variations in tritium concentration over its surface, since such a target provides a more stringent comparative test of two methods of mapping.

Figure 4 is a contour map which was obtained by X-ray mapping. This target originally contained 21×10^{18} tritium atoms/cm^2 (based on original strength of 5.2 curies), and showed only slowly varying concentrations of tritium over its surface. The numbers on the contour lines represent tritium concentration after bombardment. The beam spot in this case was largely confined to 1 quadrant of the target. Numbers on the contour lines ($\times10^{18}$) represent the tritium concentration in atoms/cm^2. The concentration increases very rapidly outside the contour marked "17," reaching the original concentration in a millimeter or so. In order to determine the values associated with the contour lines, it was assumed that the X-ray intensity from a given spot on the target was directly proportional to the tritium concentration at that spot.

An autoradiograph of this same target (L–44) was scanned with a densitometer, and a map of isodensity lines is shown in figure 5. The original densitometer map presented contours which showed density increments of only 2.5 percent, but this was too close for the present purpose. Every third line of the original contour lines is shown in figure 5, so that separations between them represent 7.5 percent changes in tritium content. Normalization was accomplished by assigning to the outermost contour the value 21×10^{18} tritium atoms/cm^2 and assuming a linear relationship between densitometer contours and tritium concentration over the map where variations are about 33 percent (14.6 to 21).

Although no attempt was made to look at fine details in mapping with the Ti K X rays, the correlation between figures 4 and 5 is quite satisfactory. The drop in tritium concentration from its original concentration (before target bombardment) is almost identical in the two maps, and the contour lines certainly show the same general tendencies. It is apparent that autoradiographs can yield detailed and satisfactory mappings of tritium targets.

Correlation Between T(p,n) Mapping and Autoradiographs

These two methods of mapping were compared by measurements on another used target (designated as L–97). Figure 6 shows contours of equal tritium concentration which were obtained by the

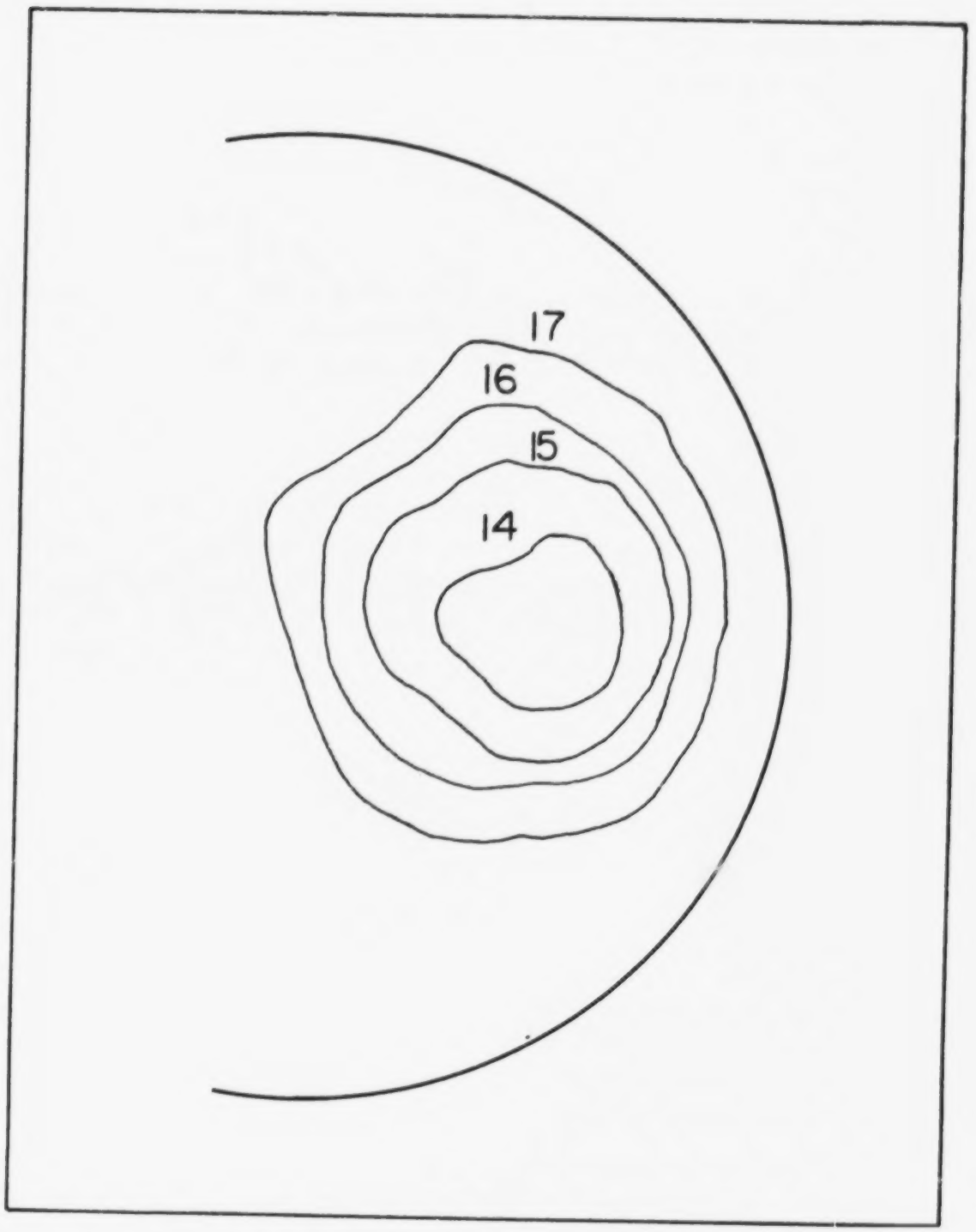

Figure 4. Map of tritium concentration as it appeared in a target (L–44) after bombardment. Numbers on the contour lines represent tritium concentrations in atoms/cm^2 ($\times 10^{18}$). Concentration before bombardment was 21×10^{18} atoms/cm^2. This map was made by observation of the T/Ti K X rays from various areas of the target. Apparatus shown in figure 2 was used for measurements. Active target diameter is 1 inch.

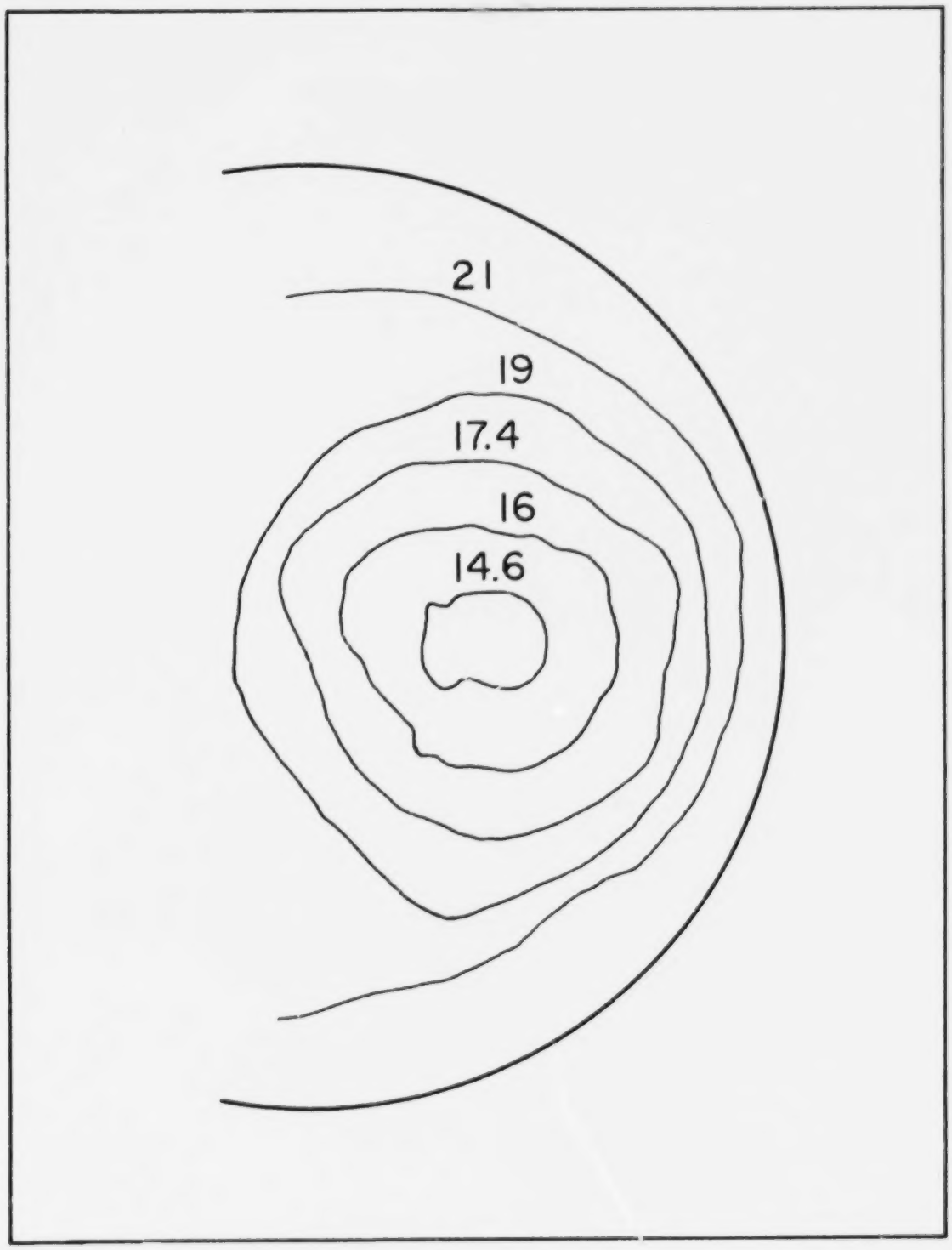

Figure 5. Densitometer scan of autoradiograph produced by the tritium decay from target L–44. The correlation between the two maps of this target is satisfactory. Discrepancies lie well within variability in tritium concentration in the target prior to its bombardment. Active target diameter is 1 inch.

T(p,n) mapping procedure. As previously pointed out, this procedure yields absolute results directly. Inasmuch as the target was mapped in only 25 positions, the contour lines shown are necessarily approximations made by visual inspection of the variations in the mapped points.

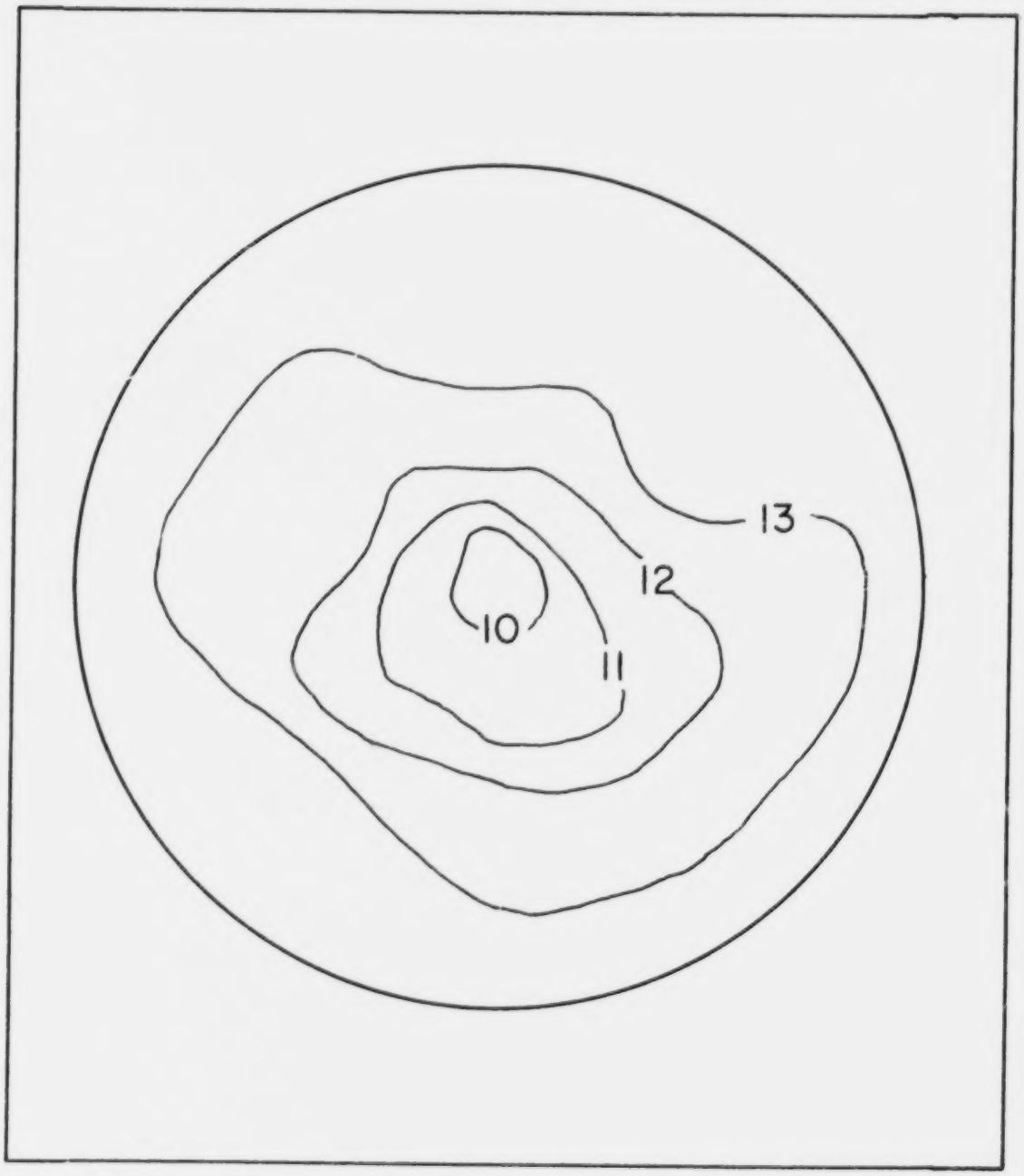

Figure 6. Map obtained for target (L–97) after its bombardment in a neutron generator. The target was removed from the generator and placed in a Van de Graaff accelerator, where it was bombarded with protons. On the basis of the yield from the T(p,n) reaction as observed in 25 different elements of area of the target, the above map was drawn. The T(p,n) reaction yields an absolute measurement of tritium concentration. Active target diameter is 1 inch.

The autoradiograph of this same target (L–97) was scanned in a densitometer, and every third contour is shown in figure 7. Variations between contour lines, as shown in figure 5, are 7.5 percent. The outer contour line was normalized to a value of 13×10^{18} tritium atoms/cm^2, and the other numbers appearing on the contour lines were then calculated. Comparison of figures 6 and 7 shows that in each map the tritium concentration falls off in the same general

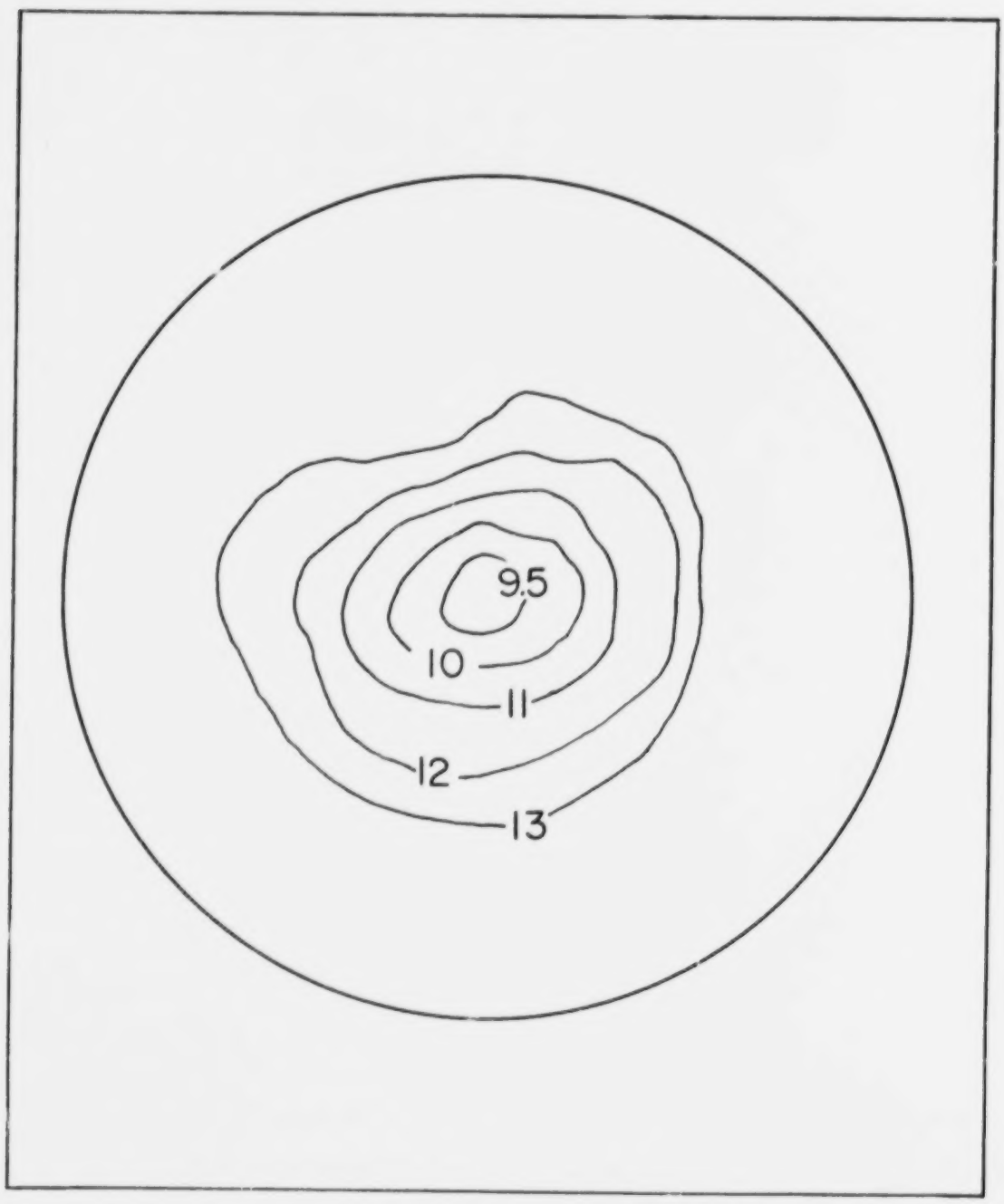

Figure 7. Map of target (L–97) reproduced from a densitometer tracing of an autoradiograph. The absolute concentration of tritium was normalized to 13×10^{18} atoms/cm^2. Correlation between the results presented here and in figure 6 is satisfactory if one considers the relatively small number of points used in the T(p,n) mapping. Active target diameter is 1 inch.

way and over essentially the same range (about 30 percent). In the light of these results and of those presented previously, it is clear that there is satisfactory correlation (within an estimated experimental error of about 10 percent) between the mapping of tritium targets by the three methods described.

B. Bombardment of Targets

After the techniques of mapping tritium targets had been established (sec. I.A.), it was possible to make quantitative studies of the loss of tritium from targets which were subjected to controlled bombardments with deuterons obtained from a Cockcroft-Walton neutron generator. Some of the maps of such targets have already been presented in the preceding section. Details of several bombardments and the results obtained are given below.

Three targets were selected and mapped by the T(p,n) technique before bombardment. During bombardment, each target was contained in the assembly shown in figure 8. This particular assembly is conventional except for the inclusion of a small cylinder through

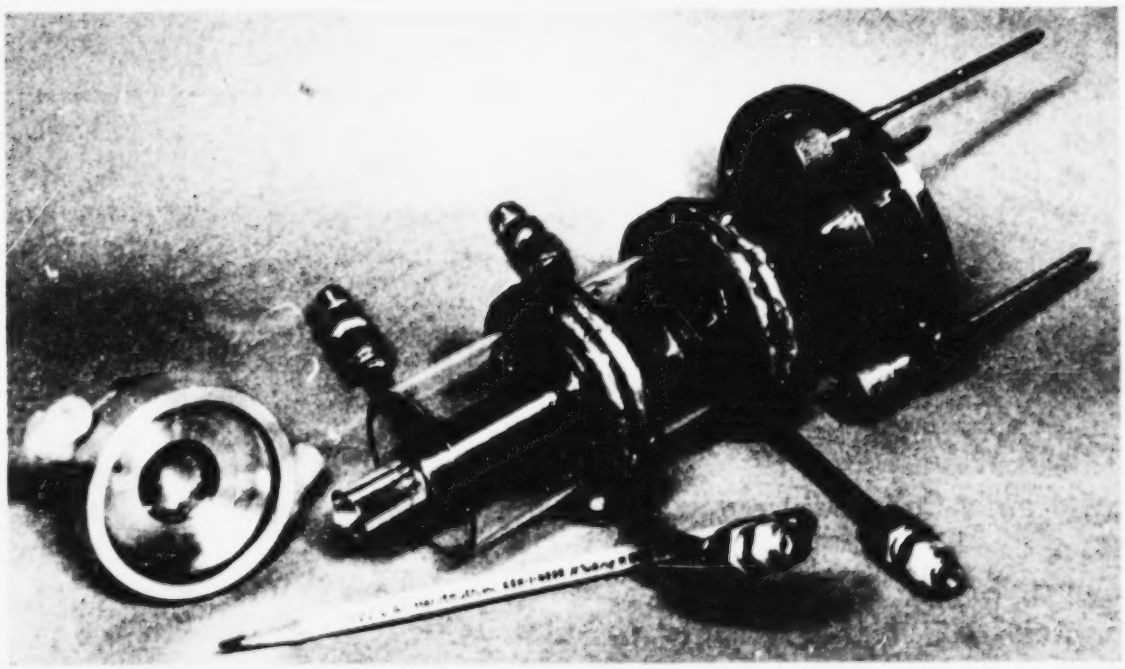

Figure 8. Photograph of the target assembly used in the "catcher" experiments. The target holder, left, shows the circular cavity through which water circulates in order to cool the target (removed when this photograph was made). The cylindrical aluminum catcher has been pulled out of its normal nesting position inside the more prominent metal cylinder. Collimators and cooling coils are shown at center right in the picture. During target bombardment nearly all sputtered material should make its first collision with the catcher.

which the bombarding beam passed and which served as a "catcher' for any titanium that might be sputtered from the target. (This point will be discussed in detail in sec. I.D.) Measurements were made of the bombarding current and of the T(d,n) neutron yield as a function of time. In this particular set of runs, the beam current was magnetically analyzed so that either the monatomic or diatomic beam could be selected for bombardment. Two bombardments were made with a monatomic beam and one with a diatomic beam. After bombardment each target was removed from the generator and again mapped by the T(p,n) technique. (Autoradiographs were also made but were not analyzed in detail except for target L–97. The results were used in the correlation studies only, as previously described in sec. I.A.)

The results of these bombardments are shown in the first three rows of table 3 (targets L–96, L–97, and L–98). The fourth row shows results obtained previously in a bombardment, with an unanalyzed beam, of target L–44. This bombardment was made before the target assembly had been completed (fig. 8).

Table 3. Loss of tritium activity resulting from bombardment of solid targets

Target number	Number of tritium atoms in target ($\times 10^{19}$)	Charge delivered (coulombs)	Type beam	Total neutron yield ($\times 10^{13}$)	Percent drop in neutron yield	Loss of target activity (mCi)
L–96	7. 63	1. 57	Molecular	. 50	30	77
L–97	6. 32	2. 45	Atomic	4. 0	42	193
L–98	9. 13	4. 96	Atomic	7. 7	20	170
L–44	[a] 10. 7	2. 7	Unanalyzed	8. 8	65	[b] 300

[a] Values based on tritium uptake. Other values in this column are results of T(p,n) mapping.
[b] Based on mapping by Ti K X-ray technique. Other values in this column are results of T(p,n) mappings made before and after bombardment.

It is evident from the data in table 3 that T/Ti targets lose significant amounts of tritium during bombardment. In these particular runs, the loss averages about 70 mCi/coulomb of bombardment. The molecular beam carries 2 deuterons/unit charge, but the effective bombarding energy is only one-half of the accelerator potential. Results on target L–96 are therefore not necessarily comparable to results involving the other targets; however, the loss for L–96 is quite comparable to the other losses observed.

In normal usage, a target is bombarded until the neutron output from it has decreased by at least 50 percent (that is, through 1 "half-life"). It is clear from the data in table 3 (note columns 3 and 6) that several coulombs must normally be delivered to a target before it

deteriorates to 50 percent effectiveness. Our data show that the target will probably give up several hundred millicuries of tritium during this period. This amount of tritium could be hazardous to personnel (unless it is properly controlled).

C. Review of Target Performance

It has been known for many years that the neutron yield from the T(d,n) reaction in tritium-metallic targets decreases with time. This fact was pointed out in the above discussion of the results shown in table 3. Previous studies of target performance have been made by Akers and Banta (*1*) and by personnel at Texas Nuclear Corp. (*3*).

A standard test for the performance of tritium targets was established by Akers and Banta (*1*). They used an accelerating potential of 150 kV, bombarding current of 500 μA, and a beam area of 0.5 cm^2, and found that targets deteriorated (with respect to neutron yield) more rapidly during the first hour of bombardment than in subsequent periods. The half-life for decay was therefore established during the second hour of bombardment when the decay rate was relatively constant. From their observations, a "figure of merit" for target performance was established. This was defined through the relationship:

$$\text{Figure of Merit} = Y_0 R_1 T_2 \qquad (1)$$

where Y_0 is a normalization constant, R_1 is the decay ratio of neutron yield during the first hour, and T_2 is the half-life during the second hour of bombardment. The "first hour half-lives," deduced from values of R_1 between approximately 0.3 and 0.6, correspond to half-lives of 35 to 80 minutes. The values of T_2 vary from approximately 50 to 100 minutes. In rough approximation, the half-lives observed (whether during the first hour or subsequent hours) are about 1 hour. This value is directly related to the current-density of their bombarding beam (1 mA/cm^2).

The tests made at Texas Nuclear Corp. (*3*) also showed the usual exponential decrease in neutron output from a bombarded target. These tests were performed on different types of tritium-metallic targets and with varying current-densities. When it became clear from the work reported above (sec. I.B.) that tritium was definitely driven from targets, it seemed that a figure of merit (M) might be defined as:

$$M = \frac{\text{number of deuterons delivered to target}}{\text{number of tritium atoms lost from target}} \qquad (2)$$

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This definition implies that a target which has a high figure of merit will lose relatively less tritium during delivery of a fixed charge of deuterons. As a target deteriorates, the value of M (defined above) must certainly increase: the number of deuterons delivered can increase indefinitely, whereas the tritium loss is limited by the total amount originally present in the bombarded area of the target. However, the rate of decay of the target is roughly constant over a single half-life. The number of deuterons delivered during 1 half-life is proportional to IT, where I is the bombarding current and T is the half-life. Inasmuch as the decrease in neutron output is directly related to the tritium loss from the target, it may be assumed that the bombarded area of the target loses one-half of its original tritium concentration during the bombardment. If the area of the bombarding beam is A cm^2, and the original concentration of tritium in the target is S Ci/cm^2, the amount of tritium lost in 1 half-life is ½ AS. A curie of tritium consists of 2.03×10^{19} atoms, and a coulomb of deuterons represents 6.25×10^{18} particles. Hence one may substitute in the preceding equation and obtain:

$$M=\frac{6.25\times10^{18}\,IT}{\frac{1}{2}AS\times2.03\times10^{19}} \tag{3}$$

It is usual to measure I in μA, T in minutes, A in cm^2, and S in Ci/cm^2. In such a case, one finds:

$$M=3.7\times10^{-5}\,\frac{IT}{AS} \tag{4}$$

Using this formula, M can now be calculated for various targets. It has been pointed out previously that Akers and Banta found half-lives of roughly 60 minutes for their targets; their bombarding beam was 500 μA and the bombarding beam had a cross-sectional area of 0.5 cm^2. Assuming that target strengths were 1.0 Ci/cm^2, one finds M=2.2. Work on target deterioration has also been reported by Broerse (*2*). He used 400-keV deuterons at 150 μA to bombard a target of strength 1.5 Ci/cm^2. Broerse observed a half-life of 17 hours with 60 watts of power over a 2 cm^2 area of target. From these data, one finds M=1.9. Although the bombarding energy was higher than that used by Akers and Banta, the values of M are approximately the same. Finally, data previously reported from Texas Nuclear Corp. was reviewed and M was calculated for a variety of bombarding beams and for various targets. The results are shown in table 4. The targets listed in the table represent various types of metallic backings and absorbers, and the concentrations of tritium vary greatly. Although data are included for these experimental targets, targets of "normal" loading are of primary interest in our present evaluation.

(A target for which S is quite small may well have a large value of M, but the neutron output is small, and the target would have little practical value.) Note that all targets with loadings of approximately 1 Ci/cm² show a value of M ≈ 1.

It is clear that the values for M, as defined by equation 2 and as calculated on the basis of results in three different laboratories, are close to unity. This is rather striking—it implies that each bombarding deuteron will, as a statistical average, remove 1 triton from a T/Ti target of normal loading. The data, however, do not reveal the mechanism of target deterioration. Attention is directed to this problem in section I.D.

Table 4. Calculated figures of merit for various bombarding beams and various targets

Target number	S Ci/cm²	A cm²	I μA	T min.	M	Target characteristics
1	1. 24	. 08	20	~160	1. 2	Ti/Cu.
2	. 68	. 08	20	~85	1. 2	Ti/Cu.
3	. 16	. 08	20	70	4. 0	Ti/Cu.
4	. 78	. 32	100	55	. 8	Ti/Cu.
5	. 78	. 32	100	46	. 7	Ti/Cu, Al overcoat.
6	. 29	. 32	100	55	2. 2	Er on Cu.
7	. 78	. 32	300	18	. 8	Ti/Cu.
8	. 78	. 32	300	15	. 7	Ti/Cu, Al overcoat.
9	. 46–. 78	. 32	300	[a] 18	. 8–1. 4	Ti/Cu.
10	. 29	. 32	300	[b] 14	1. 7	Er on Cu.
11	. 006	. 32	100	~110	~. 2	Sc on Cu (good).
12	. 006	. 32	100	~10	~. 02	Er on Cu (flaky).
13	. 007	. 32	100	~100	~. 16	Er on Al.
14	. 62	. 08	20	[c] 40	. 6	Y on Cu.
15	. 042	. 32	100	70	19. 0	Zr on Cu.
16	. 07	. 32	100	23	3. 8	U on Cu.

[a] Increased to T = 28 after ~30 minutes.
[b] Increased to T = 23 after ~20 minutes.
[c] Increased to T = 75 after ~25 minutes.

D. Mechanism of Loss

It was established through the work reported earlier (sec. I.A.) that the decline in neutron output from a target under bombardment is accompanied by an actual loss of tritium from the target. Any decrease in target performance that is due to the deposition of carbon on the target must be relatively small. Studies show that the decline in neutron output is roughly proportional over approximately the first

half-life to the decrease in tritium concentration on the bombarded area of the target.

Part of the tritium is obviously consumed in the T(d,n) reaction, some of it passes out of the system in the form of a gas, and the remainder (as was found in the course of the work) remains in the system in some combined form. These points will be discussed in more detail below.

LOSS BY THE T(d,n,) REACTION

It may be shown at once that the loss of tritium from a target through the T(d,n) reaction is very small. For example, for target L–44 a loss of 0.3 Ci of tritium (which is 6×10^{18} tritium atoms) was measured. During the course of deuteron bombardment, this target yielded approximately 8.8×10^{13} neutrons. Hence there were approximately 10^5 tritium atoms lost for each neutron formed in the T(d,n) reaction.

This result is typical of calculations which were made for other targets. It was concluded that the removal of tritium from a target through the T(d,n) reaction is completely negligible in comparison with other processes.

THE T/Ti K X-RAY SPECTRUM IN SYSTEM COMPONENTS

Quite early in the course of these investigations, it was observed that Ti K X rays were emitted from inside the stainless steel drift tube that was attached to a neutron generator which had been used over a period of many months. It was not unexpected that tritium would exist in the drift tube, and that characteristic X rays from iron, nickel, and chromium (the constituents of stainless steel) might be observed. However, the only characteristic X ray that was definitely observed was from titanium. The X-ray spectrum in the drift tube was essentially identical with the X-ray spectrum emitted by the T/Ti target. Even more surprising was the fact that the intensity of this characteristic X ray was almost the same at the two ends of the drift tube, which indicates that the titanium (in some form) diffuses to distances quite far removed from the target. Figure 9 shows the X-ray spectrum as it appeared at each end of the drift tube. An additional check on the identity of the X ray was obtained by taking alcohol-moistened Q-tip smears from both ends of the drift tube. The 4.5-keV Ti X-ray line was observed in both smears.

This observation led to the speculation that a tritium target may deteriorate through the sputtering of titanium from its surface. Just

why the sputtered titanium would not stick to the first surface it struck after its release could not be explained. The possibility that it was removed as a hydride seemed somewhat remote. This latter speculation had a numerical appeal as shown by the following statements. It had been concluded (sec. I.C.) that 1 tritium atom (as a rough average) is removed per bombarding deuteron. Furthermore, the sputtering ratio (atoms sputtered divided by number of incident particles) for titanium by deuterons, while not known explicitly (*4*), is probably of order unity in the region of 100 keV. At this point it appeared possible that the atoms of tritium lost by a target might be accompanied by an equivalent loss in atoms of titanium. With this possibility in mind, an experiment was designed to catch any titanium that might be sputtered from the target.

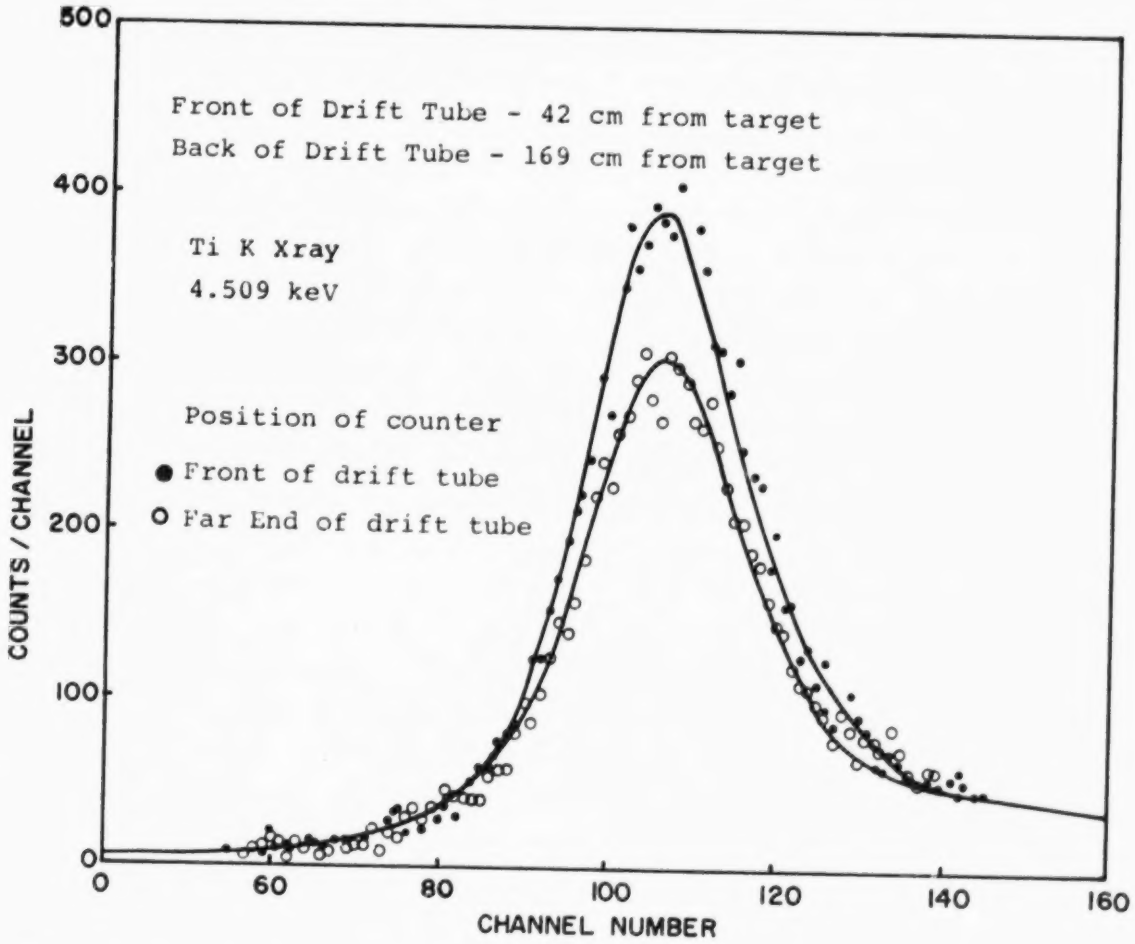

Figure 9. Spectrum of radiation obtained by counter which looked into the front end of the drift tube (that is, part nearest the target), and back of drift tube. The observed intensities differ by approximately 30 percent. The maxima appear in each case at the known Ti K X-ray position. No indication of Fe K X rays was obtained.

SEARCH FOR SPUTTERED TITANIUM

It was assumed that any titanium that might be sputtered from a target would be deposited on the first surface it struck. Accordingly, a suitable "catcher" was formed from household aluminum foil. It was a hollow cylinder 5 cm in length and 1.5 cm in diameter. This was placed inside the vacuum system, just in front of the target (fig. 8). Three sets of bombardments, each involving a new target and a different catcher, were then made. Targets were mapped before and after bombardment in order to determine the amount of tritium lost in each case. (The results of some of these mappings were also used for another purpose—viz., to determine the validity of the mapping procedures—as reported in section I.A.) Bombardments were made with analyzed beams (atomic and diatomic) and with an unanalyzed beam. The loss of activity of the targets used has already been presented in table 3. These losses range from 77 to 300 mCi/target, with an average bombardment of approximately 3 coulombs. The average loss is approximately 200 mCi, corresponding to a loss of 4×10^{18} tritium atoms.[2] If an equal number of titanium atoms were removed from the target (as speculated above), their weight would amount to 320 micrograms. The aluminum catchers were weighed before and after each target bombardment. The accuracy of the microbalance used was such that changes in weight as small as approximately 15 micrograms could be detected. (This accuracy was possible partly because of the very light weight of the catchers—their average weight was only 1.5 milligrams.) Within this limit, no change in weight of the catchers was detected in any case. It was found that the tritium activity on the catchers[3] was quite small. It varied from 0.5 to 3.9 mCi, which is less than 2 percent of the total activity lost by the various targets (table 3). It was also found that each catcher, after it had been used in a run, was emitting Ti K X rays. The yield was small but definite, although there was no opportunity to make absolute measurements of intensity.

It would be of great interest to determine just how much titanium did leave each target. It had been simply speculated—but could not be confirmed by the catcher experiments—that titanium is lost at the same rate as tritium. This loss might be determined by returning each used target to the manufacturer and having it heated and reexposed

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[2] The figure of merit for these targets is therefore $M\approx5$. This is somewhat higher than the value for targets of similar concentration (manufactured several years ago) shown in table 4, and taken at face value would indicate improved target performance. The data are too limited to be conclusive.

[3] These measurements were made by unrolling the catchers and placing them individually in an ionization chamber. The chamber details and methods of measurement are described in section II.

to a tritium atmosphere. Presumably, if no titanium has been lost from the target, the reexposed target would reveal a tritium map which would be identical to the original (preexposure) map. It is hoped that results on this point may be obtained in the near future, although the pursuit of this problem is not a part of the present study.

It will be shown later in this report (sec. II) that most of the tritium lost from a target during bombardment is actually in the form of a gas. It appears from these catcher studies that some of the gas leaves in the form of a bound titanium tritium molecule, or titanium by some mechanism gets into the system and then absorbs tritium gas. One of these assumptions must be made in order to explain the existence of the Ti K X rays which are observed in the catcher experiments and in the drift tube itself.

It is not known from this work whether the bombarding deuterons remain in the target, perhaps displacing the tritium. Broerse (*2*) does report on the bombardment of deuteride (as well as tritide) targets. Under deuteron bombardment these targets yield the so-called d–d neutrons. Broerse states, with respect to loading of a target by the bombarding deuterons, "Our experimental findings do not agree with the increase of the neutron yield which would be expected in view of the loading of the target with the deuterons of the incident beam."

The important mechanism of tritium target deterioration, from the point of view of a health physicist, is the evolution of tritium gas from a target during its bombardment. The other points presented here offer some interesting opportunities for speculation and for further experiments on secondary processes, including tritium displacement, titanium tritide formation within the system, and the loss of titanium from the target.

SECTION II. CONTAMINATION IN COMPONENTS AND IN EXHAUST GAS

On the basis of the work described in section I, it is evident that tritium leaves a T/Ti target during bombardment and that most of it is probably in gaseous form. Tritium contamination in the drift tube after several months of use has already been mentioned. It was suspected that all components of the neutron generator were contaminated, but no quantitative measures had been made up to this point. Divergent results have been reported ((*1–2*) as quoted in sec. I.) on the amount of contamination in pump oil. Boggs *et al.* (*5*) have obtained tentative data on the amount of tritium that is exhausted from a neutron generator. Three types of observations were made in the study to determine the course followed by the tritium that leaves the target. These observations were made on the static components of the generator, the pumps, and the exhaust from the forepump.

A. Contamination of Static Components

Most of the components of a neutron generator are cylindrical in shape (target holder, drift tube, connectors, accelerating tube, valves, manifolds). In order to measure the tritium contamination inside such a component, it may be made to serve as the "outer wall" of an ionization chamber. This is done by inserting an insulated rod inside the component, placing a high potential between the component and the rod, and measuring the ionization produced by the beta rays from tritium. A typical arrangement is shown in figure 10. (The electrometer and battery arrangement used in these measurements is the same as that used subsequently with an ion chamber, figure 11.)

There are few restrictions on the use of an ionization chamber to measure tritium activity, but the following precautions must be observed: (1) The ionization chamber must be of such dimensions and the gas within it at such pressure that "wall effects" are negligible. This means that the average range of the beta rays produced within

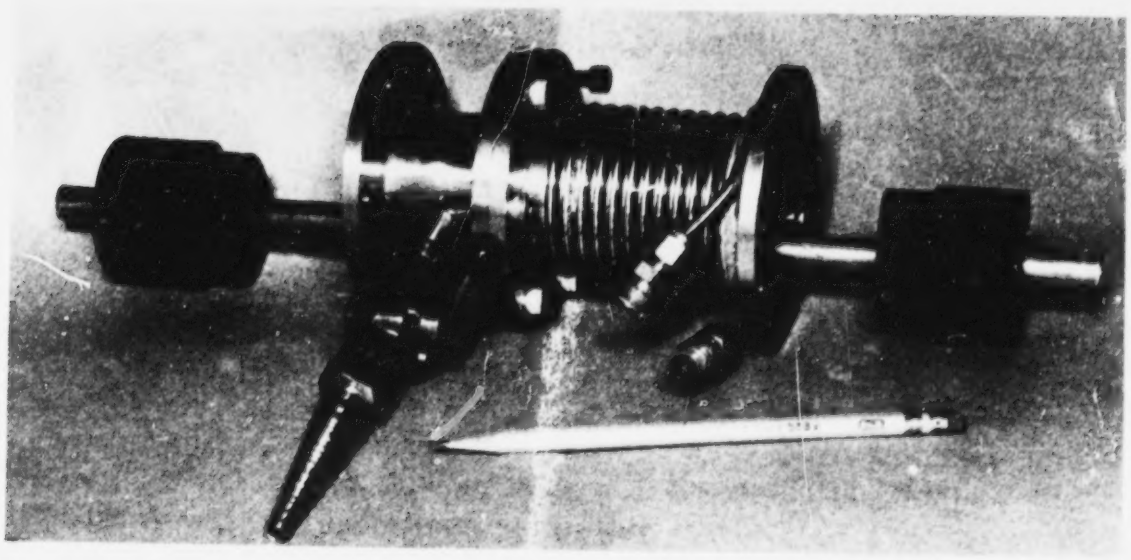

Figure 10. Illustration of method of making a part of vacuum assembly into the "outer wall" of an ionization chamber. The inner conductor of the chamber is the small rod which is held in position by the two rubber insulators.

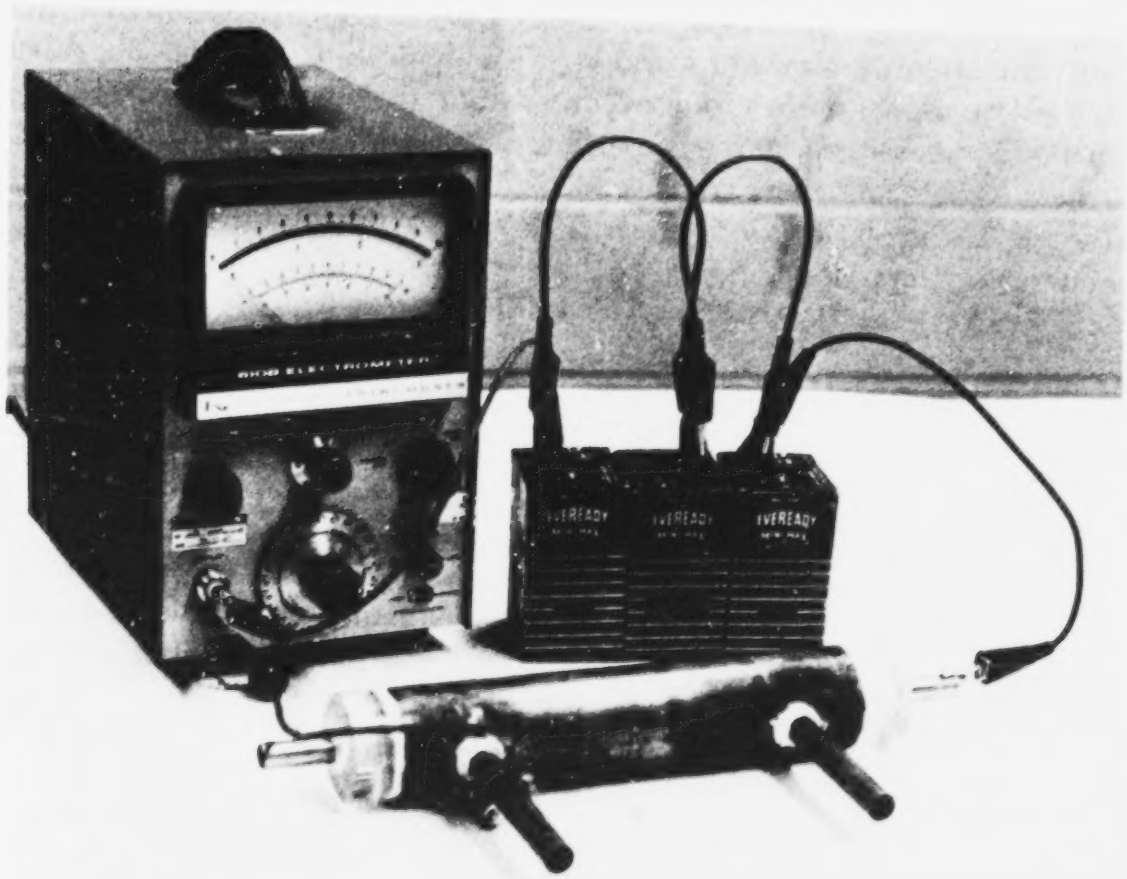

Figure 11. Ionization chamber (foreground) used to trap exhaust gas from the forepump of a neutron generator. The lucite insulators, battery connections, and electrometer complete the assembly.

the chamber must be small compared to the dimensions of the chamber itself. (2) The voltage applied to the chamber must be great enough to collect essentially all of the ion pairs which are produced by the beta rays, but it must not be so large that ion multiplication takes place. (3) Leakage and background effects must be minimized if small amounts of beta radioactivity are to be measured accurately. (4) The tritium must be in gaseous form or absorbed within a few microns of the surface of the component.

The restrictions mentioned above were all satisfied in the measurements. The components tested for tritium activity generally had minimum dimensions of several cm. (The range in air of beta rays from tritium averages only a few mm.) A map of ionization current against applied voltage showed that a definite plateau was reached at about 600 volts, and measurements were made at voltages which equaled or exceeded that amount. Leakage turned out to be small compared to the ionization current produced by tritium activity in the components. It was assumed that the metal or glass surfaces would not absorb tritium to a depth of more than a few microns. This assumption appears justified in the case of the metal parts since most of these are made of stainless steel. Iron is an endothermic occluder of hydrogen and does not readily absorb it even at elevated temperatures.

The ionization current which corresponds to a curie of tritium within a chamber is readily calculated. Assuming that the average energy of the beta rays from tritium is 5.7 keV and that the average energy required to produce an ion pair in air is 32.5 eV, one finds 175 ion pairs/beta ray. Hence the decay of 1 Ci of tritium will produce $3.7\times10^{10}\times175=6.5\times10^{12}$ ion pairs/second. One μA corresponds to a flow of 6.25×10^{12} unit charges/second. These two numbers are nearly equal, but specifically 1 Ci of tritium gives rise to an ionization current of 1.04 μA. If the tritium is absorbed on a surface, one has essentially 2π geometry, and the expected current is 0.52 μA/Ci.

The results of measurements, obtained by converting each component into the outer wall of an ionization chamber, are shown in table 5. The average amount of tritium in each of the components is in the range of a few mCi or less.

The glass section listed in table 5 was not directly suitable for ion current measurements. The conductivity was so low that even with collecting voltages as high as 1,300 volts (that is, about twice the value normally required for saturation), only about one-tenth of the expected ionization current was observed from a T/Ti target inserted in the chamber. Specifically, the maximum ionization current recorded for this target in a metal chamber was 95×10^{-9} amperes. In the glass

Table 5. Tritium contamination in components of a Cockcroft-Walton neutron generator

Component and brief description	Total ionization current ($\times 10^{-9}$ amp)	Tritium contamination (mCi)
Accelerator tube	4. 2	8. 1
Large manifold for gauge and pump connections	1. 1	2. 1
Small manifold with valve and viewer section	. 54	1. 0
Drift tube, 1⅞ inches inside diameter, 58 inches long	5. 7	11. 0
Bellows section (sylphon)	2. 1	4. 0
Gate valve near target section	. 80	1. 5
Glass section	[a]. 2	[a]. 4
"New" target assembly with "catcher" holder	. 3	. 6
Catcher holder	. 05	. 1

[a] "Normalized" to expected value for conducting material. See text for explanation.

section this value fell to 7.65×10^{-9}. The ratio of these currents, 12.4, was used as a factor to multiply the recorded ionization current for the glass section alone (that is, with the target removed). This recorded current was 1.6×10^{-11} amperes. The normalized value shown in the table is $12.4\times1.6\times10^{-11}=0.2\times10^{-9}$ amperes. This approach is admittedly open to some possible error. However, measurements made at collecting voltages intermediate to 600 and 1,200 volts showed consistent ratios of current with and without the target in the glass section, and it is believed that the value shown for tritium contamination here is essentially correct.

It should be noted that the ionization current produced by placing a tritium target in an ionization chamber is far below the value calculated on the basis of 0.52 μA/Ci of target strength. It was actually found that a target of 3.3 Ci activity produced ionization current of only 0.095 μA—or about one-eighteenth of the value which would be calculated. This "discrepancy" is readily explained by considering the form of the tritium target. The tritium itself is absorbed in a surface layer of titanium which is roughly 1 mg/cm^2 in thickness. The average range of the beta rays in titanium is only about one-tenth as great as the thickness of the titanium layer in which the tritium is absorbed. Therefore, the relatively small ionization current observed for the tritium target is readily explained to an order of magnitude. Exact thickness of the titanium layer and the problem of multiple scattering of beta rays within that layer make quantitative calculations difficult.

Inasmuch as the components described in table 5 were in use during several hundred hours of generator operation, it is estimated that some tens of curies of tritium had passed through them. The total

contamination of the static components was in the neighborhood of only tens of millicuries. It was concluded that the tritium released from the targets was either trapped in the pumps or exhausted from the system. Attention was therefore directed to these possibilities.

B. Contamination of Pumps

Particle accelerators, with the exception of the sealed tube variety, are equipped with some form of pumping system to maintain the low vacuum required for operation. The most common arrangement of pumping apparatus consists of a forepump to rough out the system, and an additional pump which is turned on after the roughing out—this produces the low vacuum required. This additional pump may be a turbomolecular pump, a diffusion pump, or an ion pump. If an ion pump is used on the system, the forepump is isolated from the system by a valve after the ion pump is started. The other pumps discharge directly into the forepump. A typical system is shown in figure 12. The degree of tritium contamination associated with three of these pumps is discussed below.

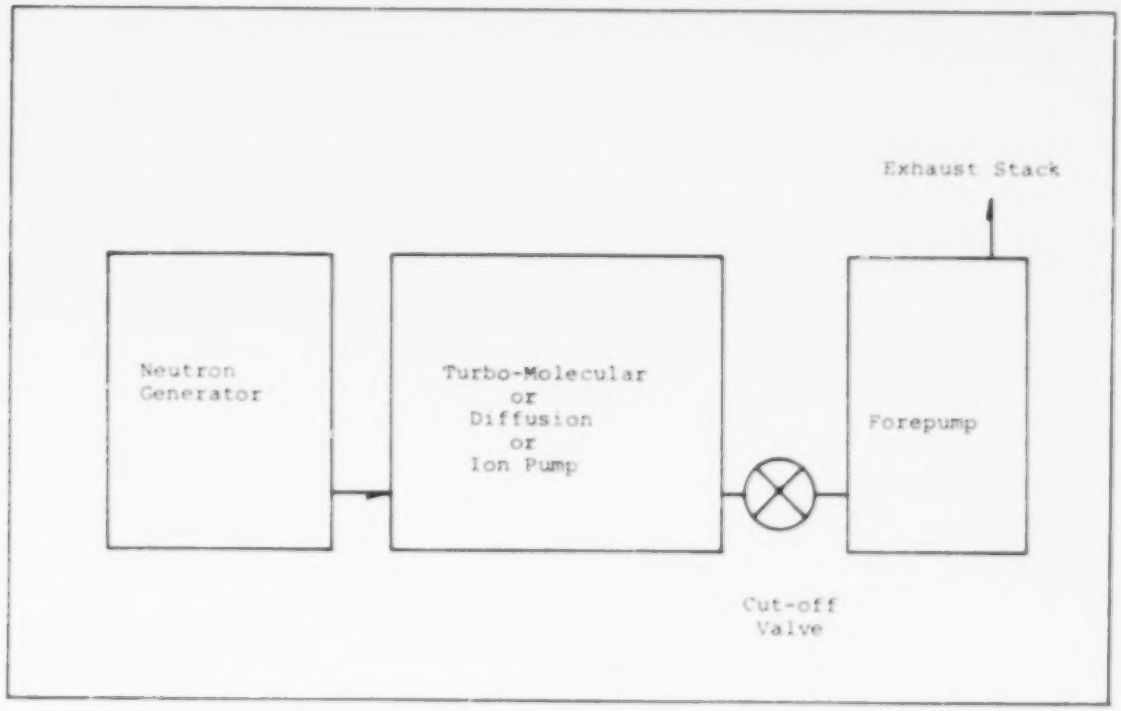

Figure 12. Block diagram of a typical accelerator pumping system. After roughing out with the forepump, the second pump is switched on. During normal operation the cutoff valve is closed when using an ion pump and open when using a turbomolecular or diffusion pump.

FOREPUMPS

Forepumps used with most systems are of the mechanical oil-filled rotary type. The air passing through the pump comes in direct contract with the oil during the pumping cycle and bubbles through a layer of oil before reaching the exhaust port. If tritium passes through the pump the logical place to look for contamination would seem to be the oil of the pump.

Samples of forepump oil from a pump that has been in continuous use for over 2 years on a Cockcroft-Walton type neutron generator were sent to two different outside laboratories for liquid scintillation analysis. The analyses were made on different dates and the following results were obtained:

Date	Tritium concentration (μCi/ml)	Total tritium in pump (mCi)
December 1966	.75	1.06
June 1967	1.08	1.53

It is estimated that some tens of curies of tritium were removed from targets under bombardment over this 2-year period, and it therefore appears that the pump oil absorbs only a small fraction of the tritium that passes through it.

DIFFUSION PUMPS

Diffusion pumps containing either oil or mercury are used between the accelerator and the forepump to obtain vacuums in the region below about 10^{-3} mm Hg. The diffusion pump is started after the pressure in the system has been lowered to approximately 10^{-2} mm Hg by the forepump.

Contamination from tritium would be expected to occur in the oil or mercury of the pump either through absorption or by attachment to the oil or mercury molecules. Accordingly, a sample of diffusion pump oil, which had been in a pump for over 2 years, was sent to an outside laboratory for analysis. The average of two measurements was:

Date	Tritium contamination (μCi/ml)	Total tritium in pump (μCi)
June 1967	1.53	610

The above results, which agree favorably with those of Broerse (*2*) (who reports 0.3 μCi/ml), indicate that the oil of these pumps does not constitute a serious tritium hazard.

ION PUMPS

In place of oil or mercury diffusion pumps, ion pumps (often called sputter ion pumps or getter ion pumps) are frequently used to produce the high vacuum required for accelerator operation. Ion pumps trap gas molecules in the pump elements, and once the pump has started its pumping action the valve between it and the forepump is closed. Basically, ion pumps consist of a stainless steel container housing one or more elements, often referred to as Penning cells. These cells consist of two titanium cathodes with a honeycomb-like stainless steel anode between the cathodes. The design of the Penning cell is such that a large number of electrons move back and forth through the anode where they ionize gas molecules that are present. The gas ions thus produced are accelerated toward the cathode, which they strike with several thousands of volts of energy, sputtering fresh titanium over the surface of the cathode. Gases like nitrogen and oxygen form stable low-vapor pressure compounds with the titanium cathode, while the light gases, such as hydrogen, diffuse into the cathode forming what is considered to be a solid solution of hydrogen in titanium.

The use of ion pumps on accelerators containing tritium can create a health hazard since the tritium is not pumped through the pump but is trapped within the pump elements themselves. In order to evaluate these hazards, several experiments were performed on used ion pumps.

Preliminary measurements on several pumps with thin end-window Geiger-Mueller counters indicated that the tritium was confined to the Penning cells (the active elements), and very little was contained in the stainless steel shells. In addition, the tritium contained in the active elements appeared to be concentrated in the titanium cathodes.

In order to obtain a quantitative value of the contamination of the stainless steel shells, liquid scintillation analyses were made on four used pump shells. The pump shells (with active elements removed) were filled with water (11 liters each) and allowed to stand for 5 days to allow the tritium in the walls to enter the water. The water samples were then sent to an outside laboratory for liquid scintillation analysis. The average of the four readings obtained was:

Date	Tritium concentration (μCi/ml)	Total tritium in water (μCi)
June 1967	2×10^{-4}	2.2

In order to determine the tritium hazards associated with the active pump elements, a large ion pump, whose general history was known, was disassembled. This pump had been operated for over 2 years on a TNC neutron accelerator, and a record of the number of tritium targets installed in the machine during this period was available. Based on the number of targets used and an estimate of the quantity of tritium removed from these targets prior to replacement, it is estimated that approximately 10 to 15 curies of tritium were removed from targets during this period.

A survey of the ion pump shell, anodes, and cathodes indicated that the tritium was confined principally to the titanium cathodes. Several of the eight cathodes were then scanned by means of a collimator and proportional counter to obtain relative yields of the 4.5-keV Ti K X ray excited by the absorbed tritium. The scans revealed a large variation in the surface tritium content over the faces of the cathodes. The concentration of tritium in the cathodes was determined by comparing these measurements with similar measurements from a titanium tritide target of known concentration. The surface measurements indicate a concentration of 1.1 Ci per pump uncorrected for self-absorption of the X ray.

In order to obtain some indication of the depth of the tritium, sections of the cathodes were milled off and the X-ray yields were measured again. It was found that the yield decreased to approximately 80 percent of the surface yield at a depth of 0.25 mm and decreased to zero at a depth of 1 mm. Based on these findings and an estimate of the self-absorption of the X ray, it is felt that the pump could contain over 10 Ci of tritium.

The active elements of an ion pump therefore do constitute a hazard if handled indiscriminately. It is recommended that when the pumps are due for repair that the Penning cells (or active elements) be replaced with new elements and the old elements be disposed of as radioactive waste.

C. Search for Tritium in Exhaust Gases

Available information on measurements of the amount of tritium expelled into the atmosphere from a neutron generator equipped with an oil diffusion pump is quite limited. Broerse (*2*) reports on a bombardment of 17 hours with 60 watts of power at 400 kV over a 2 cm^2 target. He concludes that "no more than 10^{-3} microcuries of tritium could have been discharged from the acceleration tube into the air,"

and that "most of the decrease in yield is not due to the discharge of tritium into the released gas." On the other hand, Boggs *et al.* (*5*) reached the tentative conclusion that considerable amounts of tritium were discharged into the air as a result of target bombardments and that exhaust gases from a forepump could constitute a definite health hazard unless a suitable vent or trap was included in the system.

It should be noted that Broerse apparently did not purge or vent the exhaust of his pump, but simply connected it to a tritium monitor through a rubber tube. It is conceivable that this prevented his finding greater activity. In any event, it was felt that additional measurements should be made of the activity of exhaust gases.

In order to measure the amount of tritium expelled from the system by the forepump, a small ionization chamber was constructed (fig. 12) and placed on the forepump exhaust, as indicated in figure 13. The active part of the chamber had a volume of 325 cc, and the side tubes were each of volume 25 cc. A short length of hose, with an estimated volume of 50 cc, connected the chamber to the forepump exhaust. The other side tube was connected to a capillary tube of negligible volume. An oil droplet in the capillary sealed the system and gave indication of any change in pressure within the ion chamber.

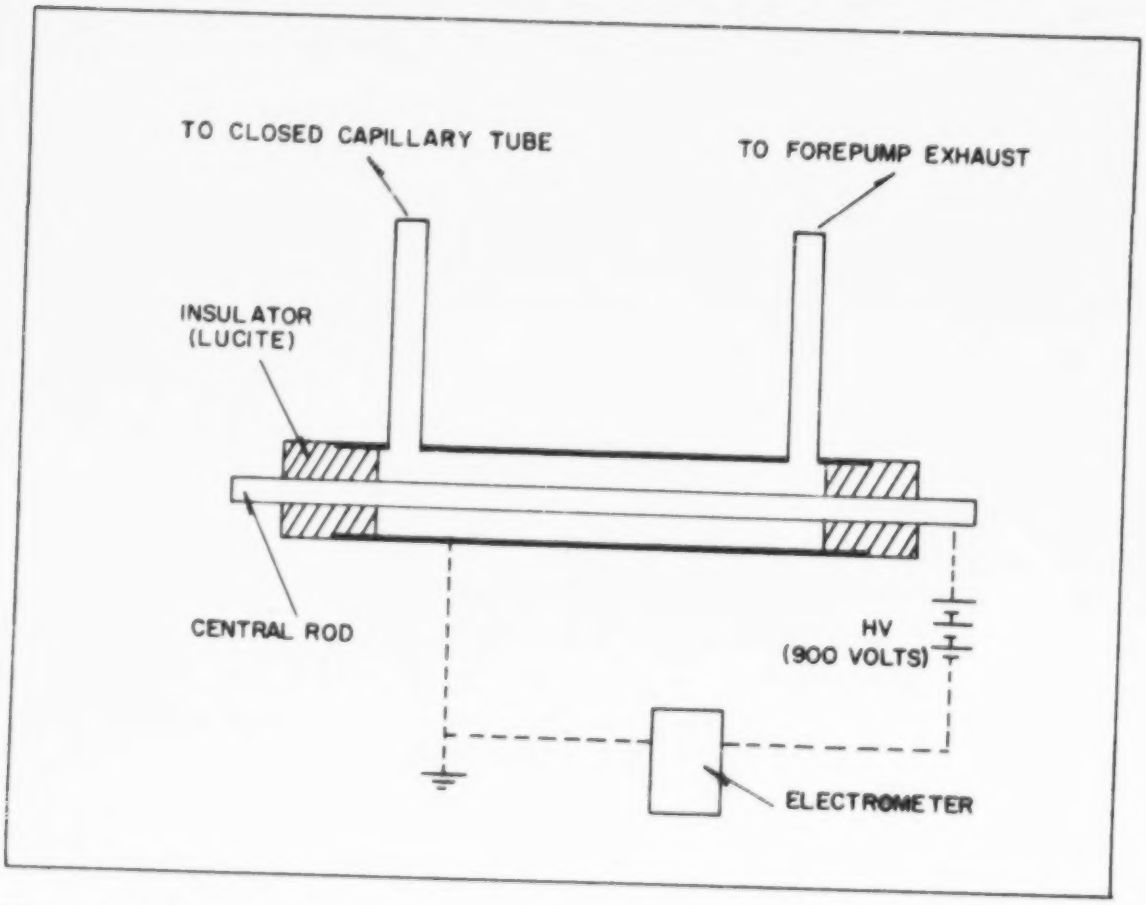

Figure 13. Diagram showing connections of the ionization chamber of figure 12 to the forepump exhaust.

The ionization chamber was initially connected to a neutron generator that had been used during the previous day. The ionization current, with the machine off, rose slowly from its background reading of approximately 4×10^{-11} amperes to 57×10^{-11} amperes in about 30 minutes. The ionization current continued to rise slowly thereafter, as shown in figure 14. There was no appreciable change in the pressure of the chamber during these observations. A change of volume of only a fraction of a cc would have been easily observable if temperature changes and/or barometric changes did not make compensation. We

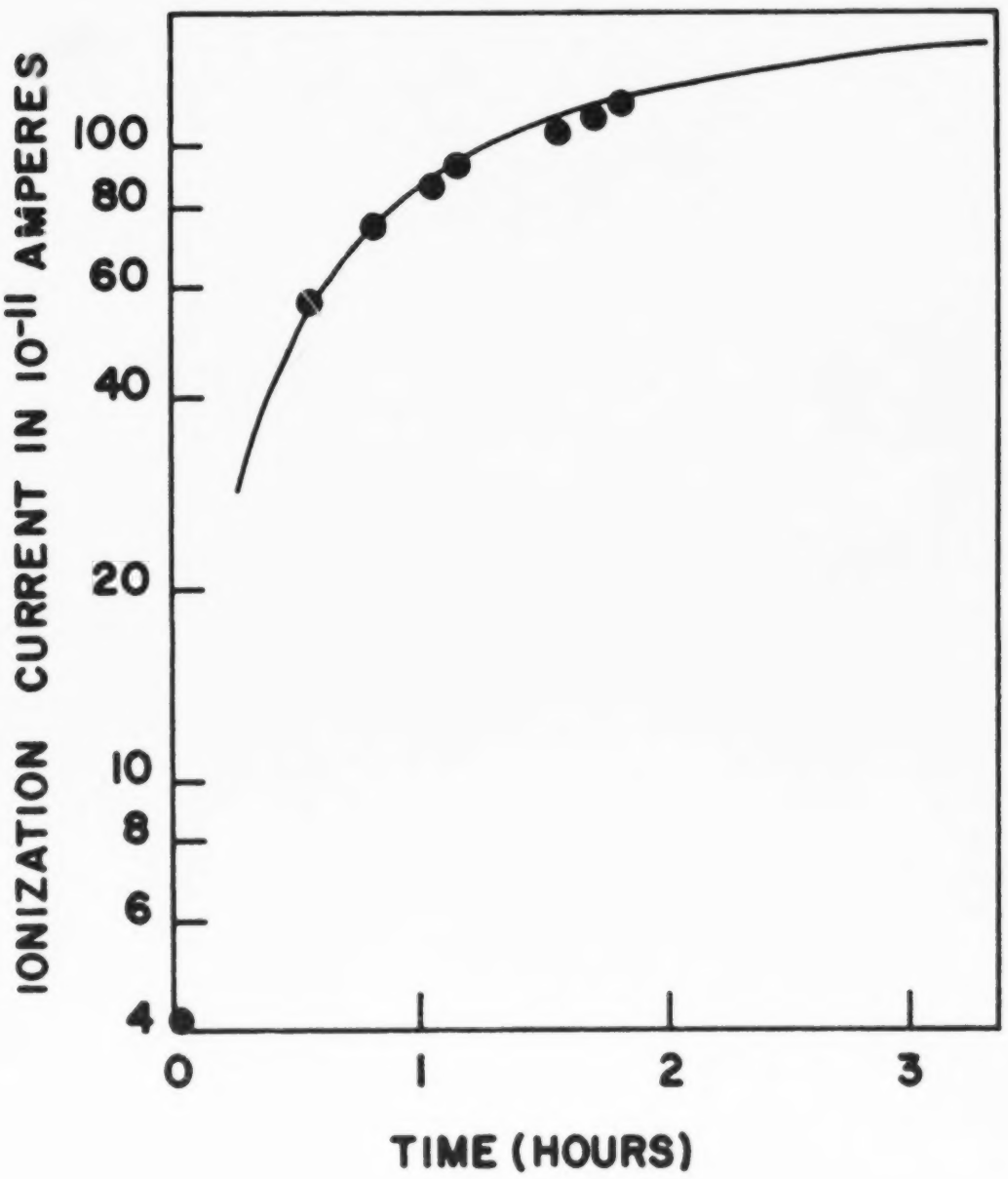

Figure 14. Increase in ionization current in the chamber shown in figure 13 as a function of time after chamber was attached to forepump exhaust. The dots represent experimental points, and the solid line is a calculated fit under the assumption that the half-life for tritium removal from the active volume of the ionization chamber is 57 minutes.

tentatively concluded that the forepump was expelling tritium which had accumulated in the system during the operation on the previous day, and that possibly it had a "residence time" of several hours or more in the pump oil.

A best fit to the experimental points of figure 14 was obtained by writing $I = K\ (1-e^{-\lambda t})$, where K and λ are constants. This is precisely the form of the buildup of a radioactive substance whose decay constant is λ. A best fit was obtained for $\lambda = 0.012$/minute, corresponding to a "half-life" of 57 minutes. It appeared that there was a regular flow of tritium into the ionization chamber, but it was being removed from the active region of the chamber by absorption or exchange—mechanisms which probably involved the rubber tubing or the lucite insulators of the chamber.[4]

The ionization current dropped overnight to a value approaching normal background. This indicates that the tritium released during bombardment was expelled by the forepump over a period of a day or so.

The following day, the target of the neutron generator was bombarded with 400 μA of 150-keV deuterons for about 20 minutes. The current in the ionization chamber rose essentially in the form already shown in figure 14. When the current was approximately 200×10^{-11} amperes, the ionization chamber was removed from the system and the side tube which had connected it to the forepump was plugged. The ionization current was now observed as a function of time, and the results are shown in figure 15. The decay of the current closely follows a simple exponential, corresponding to a half-life of about 1 hour. These data therefore correlate directly with the observations on buildup. Purging the chamber decreased the ionization current to its background value. These observations indicate that the gas in the chamber must be tritium (air bombarded by fast neutrons does not yield radioactive products of half-lives which even approximate 1 hour), and induced radioactivity in the walls of the chamber or in the room cannot be responsible for the ionization.

One final observation of tritium in the ionization chamber was made by connecting a glass cell containing approximately 1 Ci of tritium gas [5] to one of the side tubes with the other side tube plugged.

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[4] The buildup of ionization current was actually not analyzed until subsequent observations proved that the ionization current decreased exponentially when the chamber was removed from the pump, as described in the succeeding paragraphs

[5] The sample had an activity which was known only roughly, but it was used simply to obtain a qualitative check of expected ion chamber currents.

The breakoff seal was broken, and the tritium slowly diffused into the active region of the ionization chamber—a process hastened by local heating. The ionization current rose to 0.7 μA after about 20 minutes. From 1 Ci of tritium a total current of about 1 μA would be expected (sec. II.A.). However, ions could be collected from only about seven-eights of the volume of the chamber (not from the side tubes or the glass cell which contained the tritium), the tritium is absorbed within the chamber, and there are wall effects estimated at roughly 20 percent. The observed ionization current therefore lies within expectations. The current began to decline after reaching the maximum at 0.7 μA. A plot of the current as a function of time thereafter yielded a curve which was precisely parallel (within experimental error) to that shown in figure 15. It was concluded that tritium was indeed absorbed within the chamber with a half-life

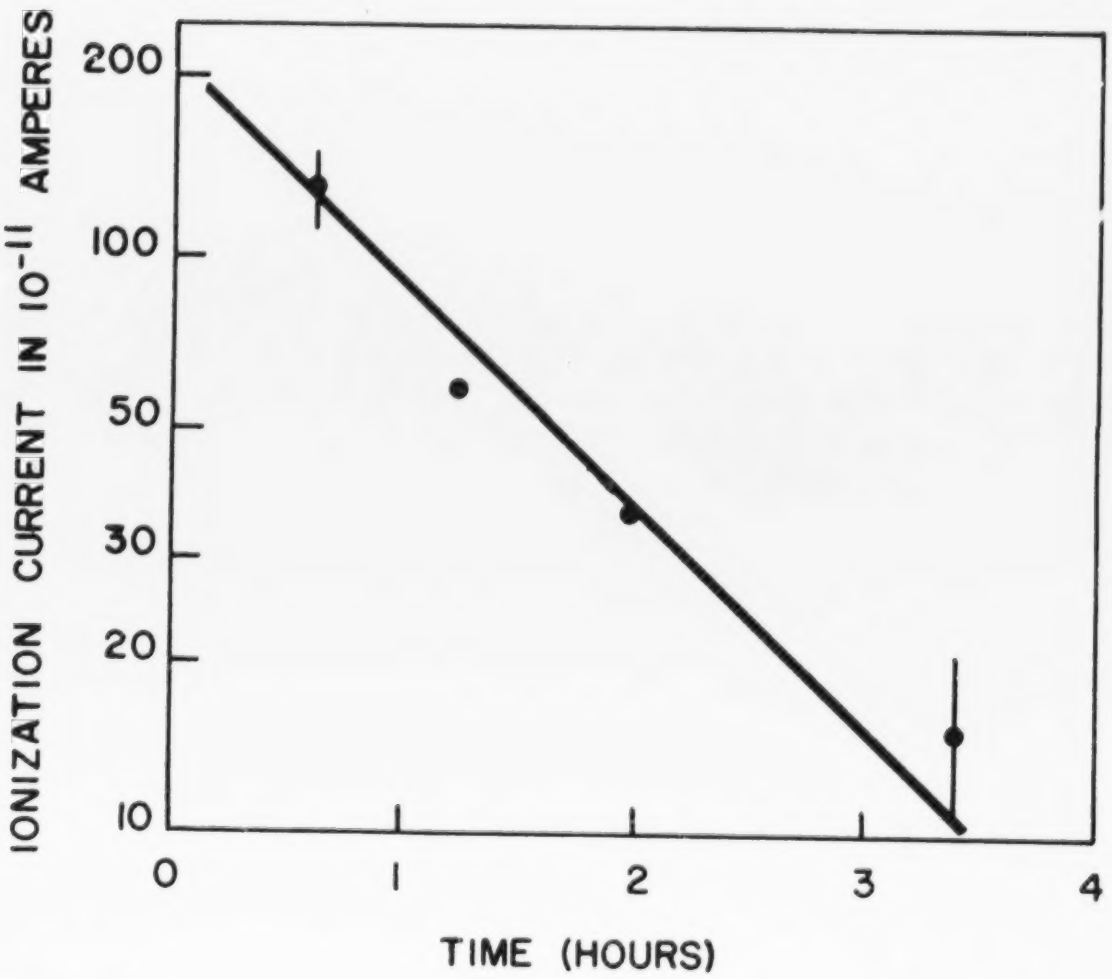

Figure 15. Map showing the decrease of ionization current with time after chamber was removed from the accelerator system, or about 1 hour after the bombardment of the tritium target had stopped. The half-life is approximately 1 hour. It is believed that the ionization current arises from tritium decay and that the observed half-life is due to the absorption of tritium in the ion chamber (probably in the insulators).

of about 1 hour, confirming the speculation that was based on the decay of ionization current produced by the exhaust gas from the forepump.

Data on ionization current as a function of time (presented in figs. 14 and 15) make it possible to estimate roughly the amount of tritium that was expelled from the forepump as a result of the target bombardment. The maximum current extrapolates to nearly 200×10^{-11} amperes. This corresponds to about 2 mCi of tritium in the chamber—perhaps as much as 2.5 mCi—after corrections are made for wall effect and inactive volume. It has already been stated that the tritium influx into the chamber continues for several hours following bombardment; 10 to 20 hours have been used as a rough estimate. These must be taken as only order-of-magnitude estimates, but indicate a tritium loss in the neighborhood of 40 mCi as a result of delivering 0.5 coulombs of charge (unanalyzed beam) to the target. This corresponds to approximately 80 mCi/coulomb. The losses reported in table 3 of section I for unanalyzed and atomic beams vary from 30 to 100 mCi/coulomb. These values also agree in magnitude with those reported by Boggs *et al.* (*5*).

The experiments reported in section I showed clearly that tritium leaves a target during bombardment. The work described immediately above shows that the tritium has a residence time of several hours in a diffusion-pumped system, but a large fraction of it is slowly expelled through the forepump exhaust. Data are too limited to prove conclusively that most of the tritium lost by a target is so expelled, but the observations made are consistent with that hypothesis.

Finally, it should be pointed out once more that the tritium was rapidly absorbed within the ionization chamber, presumably in the lucite insulators. This indicates that a plastic wool filter placed in the forepump exhaust (or elsewhere in the system) may readily remove any tritium that might otherwise be pumped into the atmosphere.

SECTION III. TRITIUM CONTAMINATION IN VAN DE GRAAFF ACCELERATORS

The problem of tritium contamination in Van de Graaff accelerators is perhaps of lesser general interest to the public than contamination in Cockcroft-Walton or sealed-tube type neutron generators. The Van de Graaff (as an accelerator for positive ions) is used primarily in research and is found principally in academic institutions and research laboratories.

Tritium is often used in gaseous form as a target material in Van de Graaff accelerators, where it is trapped in a gas cell which is sealed off from the main accelerator vacuum by a thin metal foil. The tritium is supplied to the gas cell from a uranium tritide furnace connected to the gas cell through a manifold. The furnace is heated and tritium is driven into the gas cell until the pressure gauge on the manifold indicates the desired pressure in the cell. The gas cell is then valved off and the uranium furnace cooled. At the end of the experiment, the valve is opened and the tritium gas is reabsorbed in the uranium furnace.

The hazards associated with tritium gas cells are: (1) tritium leakage into the room during filling, (2) tritium leakage into the room during operation, and (3) rupture of the metal foil.

Leakage into the room during filling is easily detected by a tritium sniffer placed near and above the gas cell. If a leak should occur, the uranium furnace is quickly cooled to prevent additional tritium escape, and the leak is then repaired. Since most accelerator rooms are equipped with good exhaust systems, the tritium leakage is not a serious problem. Tritium release into the room from a small leak during Van de Graaff operation is not easy to detect. Radiations such as neutrons and gamma rays in the vicinity of the target affect the operation of tritium sniffers and make them unreliable. The leak is usually detected as a loss in counting rate from the reaction under study and may be confirmed with a sniffer after the Van de Graaff beam has been turned off. Several hundred mCi of tritium may be released before such a leak is detected, but since tritium rises it is

soon expelled by the exhaust fans which should be on during operation. Rupture of the metal foil separating the tritium gas from the main vacuum system can occur either as a slow leak or as a burst. The tritium is released into the vacuum system and is exhausted through the pumps and out the exhaust stack. Depending upon the size of the gas cell and the tritium gas pressure used, more than 10 Ci of tritium is sometimes released in such a rupture. The contamination hazard of such a rupture is discussed below.

In addition to the gas targets, conventional solid titanium tritide targets of the type used in a Cockcroft-Walton generator are also used in a Van de Graaff accelerator. The solid target half-life is quite long when used in a Van de Graaff accelerator since the beam currents used are orders of magnitude smaller than those used in the Cockcroft-Walton generators.

Some of the hazards associated with external tritium leaks were discussed above, and attention is now turned to the hazards associated with internal rupture of gas cells and the use of solid targets. Solid targets have the same hazards associated with their use in Van de Graaff accelerators as they have in a Cockcroft-Walton generator but to a lesser degree. The actual removal of tritium is small except at high bombarding currents, as evidenced by the apparent long half-lives of targets. Some tritium is removed as titanium hydride, presumably by sputtering, but this remains close to the target region.

In order to determine the tritium contamination associated with sputtering, smears were taken with alcohol-moistened Q-tips and examined with a thin end-window proportional counter. Observation of the titanium K X ray was taken as evidence of tritium contamination. Tritium was found in the target holder, on the collimators, and on the short drift tube (plastic and metal) sections near the target. This tritium appeared to be in the form of a solid titanium tritide dust since it did yield the titanium X ray and was easily removed by thorough wiping with a moist Q-tip or by washing the contaminated part with soap and water. Quantitative values were not obtained, but by comparison with the titanium X rays from a titanium tritide target it was estimated that the contamination is in the μCi range. The contamination near the target was the greatest and appeared to be limited to a few feet up the drift tube from the target. Smears taken from the internal beam chopping slits, some 15 feet from the target, showed no evidence of tritium.

In order to assess the problem of contamination because of a gas cell foil rupture, an oil sample from a Van de Graaff forepump was sent to an outside laboratory for liquid scintillation analysis. The oil in the pump had been in use for several months, and the pump had been in operation during a foil rupture some 10 days prior to taking

the oil sample. It is estimated that approximately 3 to 4 Ci of tritium were released suddenly in this rupture and passed through the oil pump. The results of the analysis were:

Date	Tritium concentration (μCi/ml)	Total tritium in pump (mCi)
December 1966	.78	1.1

On the basis of this analysis it would appear that the gaseous tritium passes right through the pump oil with little or no absorption.

Shortly after this the oil in the pump was replaced by fresh oil, and the old oil was disposed of as radioactive waste. After 5 months and over 1,000 hours of operation, much of which involved the use of tritium targets, additional oil samples were taken. In addition to the regular forepump sample, an oil sample was taken from an auxiliary pump which is used to pump out the target section during target changes. The results were:

Date	Pump	Tritium concentration (μCi/ml)	Total tritium in pump (μCi)
June 1967	Regular	9.0×10^{-2}	127
June 1967	Auxiliary	2.8×10^{-2}	40

On the basis of the above it would appear that use of tritium either in gaseous form or as a solid tritide target does not constitute a serious hazard provided appropriate precautions are taken. Solid targets and uranium furnaces for making gas targets have been in use on the Van de Graaff accelerator at Texas Nuclear Corp. for over 8 years with no serious contamination problem. The Los Alamos group (*6*) has had similar experience.

SECTION IV. SEALED-TUBE NEUTRON GENERATORS

Compact sources of fast neutrons of energy near 14 MeV are available as sealed-tube neutron generators. These sources have found application in research, oil well logging, and activation analysis. In general, the generator consists of a sealed metal case—a few inches in diameter and 2 to 3 feet long—which contains a conventional tritide target and an ion source. The accelerating voltage (generally 100 to 200 kV) and ion source control are supplied to the ends of the case through external cables.

Unlike the Cockcroft-Walton type generator, the sealed tube has no continuous external gas supply to feed the ion source; instead, the sealed tube contains a replenisher consisting of gas contained in a titanium foil wrapped around a heating element. Variation of the voltage across the heating element releases sufficient gas from the replenisher to maintain the plasma discharge in the ion source.

The lifetime of sealed tubes, with conventional tritium targets and with pure deuterium gas in the tube and replenisher, is limited by the lifetime of the target. In order to improve the lifetime of such tubes, some designers use a mixture of tritium and deuterium in both target and ion source replenisher. This apparently extends the lifetime of the target by replenishment of the tritium which is driven out of the target during bombardment (*7–8*).

The tritium hazard associated with accidental breakage of such sealed-tube generators is small even if the breakage occurs during operation of the generator. A survey of a number of sealed-tube neutron generators indicates that these tubes contain 5 to 10 curies of tritium, but the greatest proportion of this is occluded in the target or replenisher. This can be shown by the following analysis. By virtue of the tube design (*7–8*), high-voltage breakdown in the tube occurs if the pressure exceeds approximately 2×10^{-2} mm Hg. The internal volume of a typical sealed tube is approximately 2 liters, which at

the above pressure is equivalent to a volume of 5.25×10^{-2} cm^3. There are 2.59 Ci of tritium/cm^3, so the above volume represents:

$$\left(2.59\frac{\text{Ci}}{\text{cm}^2}\right)(5.25\times10^{-2}\ \text{cm}^3)=103\ \text{mCi}.$$

The gas used in these tubes generally consists of a 50–50 mixture of tritium and deuterium. If the tube should break, a release of some 65 mCi of tritium gas can occur. In a 10- by 10-foot room with a 10-foot ceiling this could result in a tritium concentration of 2.4×10^{-3} μCi/cm^3.

The occluded tritium remaining in the broken tube presents no more than the normal hazard involved in handling conventional tritium targets. The tube should be kept away from sources of heat, which might release additional tritium, and should be disposed of as radioactive waste as soon as possible.

SUMMARY

1. Target deterioration of T/Ti targets is brought about by an actual loss of tritium during the course of bombardment (in either conventional neutron generators or in Van de Graaff accelerators). Most of the tritium is in gaseous form when it leaves the target, but Ti K X rays produced by beta rays from tritium were detected in places both near and remote from the target. This indicates that a small fraction of the tritium remains in the system in the form of a "tritide."

2. The tritide mentioned above constitutes a hazard since it readily adheres to fingers that might touch the internal surfaces of accelerator components. (This hazard may be minimized by simply washing the internal surfaces of accelerator components with alcohol-moistened cotton.)

3. The mapping of tritium concentration in a target through densitometer scans of autoradiographs and through observation of Ti K X rays yields results that are in agreement (within 10-percent experimental error) with mapping obtained by observation of the T(p,n) reaction in targets bombarded by 2.7-MeV protons. This latter type of mapping is absolute in nature, and integrated results from it yield values for tritium content which agree with those obtained (at time of target manufacture) by measurement of tritium absorption.

4. Targets of T/Ti lose roughly 1 tritium atom per bombarding deuteron. This corresponds to a loss of approximately 0.3 Ci/coulomb.

5. A generalized "figure of merit"—applicable under varying conditions of bombarding current, cross-sectional area of beam, and tritium concentration of the target—is defined and discussed in relation to paragraph 4 above.

6. Attempts to catch titanium removed from a target by deuteron bombardment yielded no positive results. Some titanium must be lost (refer to paragraph 1), but the amount appears to be small compared to the amount of tritium that is removed.

7. Typical components (except for pumps) of a neutron generator showed only minor tritium contamination (a few mCi or less) after some tens of curies of tritium have been removed from bombarded targets and passed through the vacuum system.

8. There is evidence that tritium released from the target has an average residence time of several hours in oil pumps (diffusion or forepump) and is expelled by the forepump exhaust. Measurements on a system containing an ion pump (rather than an oil diffusion pump) indicate that most of the tritium lost by a bombarded target is trapped in the elements of the pump.

9. In certain ionization chamber observations, it was found that tritium was removed rather rapidly from the active volume of the chamber. This removal is presumably due to absorption or exchange in the lucite insulators of the chamber. The results indicate that a filter of lucite wool would be a simple, inexpensive, and very effective trap for tritium gas.

10. Essentially all of the gaseous tritium released into a Van de Graaff accelerator by accidental rupture of a tritium gas target is vented from the forepump exhaust.

11. Accidental breakage of a sealed-tube neutron generator can release between 50 and 100 mCi of tritium. In a well-ventilated room this is not a serious hazard.

REFERENCES

(*1*) Akers, I. K. and H. E. Banta. Solid tritium target quality studies. ORINS Internal Report, Oak Ridge, Tenn. (1963).

(*2*) Broerse, J. J. Effects of energy dissipation by monoenergetic neutrons in mammalian cells and tissue. Publication of the Radiological Institute TNO of the Organization for Health Research, Rijswijk (Z. H.) The Netherlands (1966) pp. 23–40.

(*3*) Morgan, I. L. Tritiated targets of Sc, Ti, Zr, Er, Y, and U. Presented at the Second International Meeting on Accelerator Targets Designed for the Production of Neutrons. Grenoble, France (June 1965).

(*4*) Almen, O. *et al.* Eighth National Symposium on Vacuum Technology I:247 (1961).

(*5*) Boggs, R., R. Jacobs, and R. L. Leupold. Radiological health problems associated with the operation of neutron generators. Presented to the Eleventh Annual Meeting of the Health Physics Society, Houston, Tex. (June 30, 1966).

(*6*) Engelke, M. J. Proceedings of the Symposium on Instrumentation, Experience and Problems in Health Physics Tritium Control. Sandia Corp., Albuquerque, N.Mex. (February 16–17, 1967).

(*7*) Reifenschweiler, O. Neutrons from small tubes—Phillips tube: continuous or pulsed operation. Nucleonics 18:69 (1960).

(*8*) Bounder, J. E., P. D. Lomer, and J. D. L. H. Wood. A neutron tube with constant output (10^{10} n/sec) for activation analysis and reactor applications. Nucl Instru Methods 33:283 (1965).

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